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Title: Ionizing Radiation Processing of Fruits and Fruit Products

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Citation: In "Processing Fruits: Science and Technology, Second Edition" Publisher by CRC Press Boca Raton, FL 2005 Chapter 11, Page (2005) 223-261

Number: 7443

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11 Ionizing Radiation Processing of Fruits and Fruit Products

B.A. Niemira and L. Deschenes

CONTENTS

11.1	Introduction	224
11.2	Ionizing Radiation Physics and Technologies	225
11.2.1	Gamma Rays	226
11.2.2	Accelerated Electron Beam (E-Beam)	227
11.2.3	X-Rays	227
11.2.3	General Mode of Action	228
11.3	Chemical Effects of Ionizing Irradiation in Foods	229
11.3.1	Macromolecules	229
11.3.2	Small Molecules	229
11.3.3	Protection in Complex Foods	229
11.4	Biological Effects of Ionizing Irradiation in Foods	229
11.4.1	Principal Targets	229
11.4.2	Sensitivity of Organisms	230
11.5	Technical Aspects of Food Irradiation	230
11.5.1	Dosimetry	230
11.5.2	Detection of Prior Irradiation	231
11.6	Irradiation Facilities	232
11.7	Controlling Regulations: Commodities, Dose Limits, and Purposes	234
11.8	Applications of Irradiation to Fruit	235
11.8.1	Delay of Ripening	236
11.8.2	Disinfestation	236
11.8.3	Reduction of Microbial Load	236
11.9	Response of Fruit Tissue to Radiation Treatment	237
11.9.1	Wound Response	237
11.9.2	Delay of Ripening	238
11.9.3	Postharvest Disease and Storage Losses	238
11.9.4	Physiological Disorders	239
11.10	Quality of Irradiated Fruits	239
11.10.1	Texture, Color, and Sweetness	239
11.10.2	Flavor and Aroma	239
11.10.3	Nutritional Quality	240
11.10.4	Influence of Variety/Cultivar	241
11.10.5	Quality Control	241
11.11	Irradiation of Fruit Juices and Pulps	241
11.11.1	Microbiology of Irradiated Juices and Pulps	241
11.11.2	Sensory Properties of Irradiated Juices and Pulps	242

11.12 Fruits Currently Being Irradiated	244
11.12.1 Strawberry	244
11.12.2 Mango	245
11.12.3 Papaya	245
11.13 Combination Treatments	246
11.13.1 Mild Heat	246
11.13.2 Modified Atmosphere Packaging (MAP)	246
11.14 Irradiation of Packaging Material	247
11.14.1 Types of Plastics Used in Fruit Packaging	248
11.14.2 Effects of Irradiation on Packaging Plastics	249
11.14.3 Regulation of Packaging Materials for Use with Irradiated Products	250
11.15 Cost-Benefit of Fruit Irradiation	250
11.16 The Future of Irradiation Processing of Fruits	252
Acknowledgments	253
References	254

11.1 INTRODUCTION

Food irradiation is a physical treatment in which food is exposed to ionizing radiation, i.e., radiation of sufficient energy to expel electrons from atoms and to ionize molecules. This radiation may be in the form of high-energy photons (gamma rays or x-rays) or accelerated electrons in the form of an electron beam (e-beam). Foods treated with ionizing radiation have consistently been shown to be wholesome and nutritious [1–9]. Numerous reviews have been written on food irradiation [10–14], including reviews that specifically address the irradiation of fruit and vegetable products [15–17]. Despite the extensive body of evidence and the virtual consensus among researchers, acceptance of irradiated foods by the general public has historically (i.e., pre-2001) been comparatively low [18–20]. This reluctance had been ascribed primarily to a failure on the part of food scientists and food safety experts to adequately educate consumers about the benefits of food irradiation and to dispel persistent fallacies about the process, such as the myth that irradiated food becomes radioactive [21]. Frenzen et al. [22] found that 45.9% of 10,767 U.S. adults surveyed had never heard of food irradiation. Irradiated meats, fruits, and vegetables have been commercially available in the U.S. since 1992, although in limited markets. In 2000, food processors in the U.S. began a major effort to introduce irradiated ground beef and papayas to the marketplace, which led to a steady growth in the capacity of food irradiation facilities in the U.S.

In October 2001, following bioterrorist attacks in which spores of *Bacillus anthracis* were mailed to public figures in the U.S., the U.S. Postal Service began using e-beam technology to irradiate letters and parcels to eliminate bacterial spores. For several weeks, the benefits and technical details of the application of ionizing radiation to a consumer product as an antimicrobial measure were discussed at length in a variety of broadcast, print, and Internet news outlets. As a statement of the safety of the process, the reports typically cited the use of irradiation on food, and usually quoted assessments from the Centers for Disease Control and Prevention, which said that irradiation is a safe and effective technology that can prevent many foodborne diseases, [2] and from the U.S. Food and Drug Administration (FDA), which determined that the process is safe and effective in decreasing or eliminating harmful bacteria [7]. This media exposure provided an unprecedented public educational program. A poll of 1008 U.S. adults conducted in November 2001 showed a markedly increased level of support for food irradiation — with 52% of the participants agreeing that the government should require irradiation of foods to ensure a safe food supply — as compared to only 11% of consumers polled in 2000 who stated a willingness to buy irradiated foods [23].

This shift in consumer attitude toward irradiation as a tool to increase the safety of consumer products is mirrored by a change in the way irradiation is regarded by food processors and researchers. Rather than as a tool to extend product shelf life, much of the latest research on food irradiation, particularly with regard to irradiation of fruit and vegetable products, has been on the elimination or attenuation of food-borne pathogens such as *Escherichia coli* O157:H7, *Listeria monocytogenes*, and salmonella [16, 17, 24, 25]. Early studies with irradiated produce typically used relatively high doses to completely eliminate spoilage bacteria and fungi [26]. The doses employed often exceeded the maximum radiation tolerances of vegetable commodities tested, resulting in loss of quality [27, 28]. Irradiation was, therefore, generally regarded as unsuitable for application to fresh produce [29–31]. However, low dose irradiation, i.e., less than 3 kGy (0.3 Mrad), is currently seen as one of several potential sanitary approaches to fruit and vegetable processing [16, 25, 32]. Recent research has shown that bacteria may be internalized in fruits and vegetables beyond the reach of surface sanitizers [33, 34]. The penetrability and subsurface antimicrobial efficacy of irradiation suggest that it can play an important role in the sanitization of produce.

AU: "Hurdles" are obstacles, negatives; change OK?

The United Nations Food and Agriculture Organization (FAO) and International Atomic Energy Agency (IAEA) recently initiated a cooperative research program on the use of irradiation to ensure hygienic quality of fresh, pre-cut fruits and vegetables and other minimally processed food of plant origin [35]. This program brings together food scientists from 15 nations, including Brazil, Canada, China, India, the U.K., and the U.S., to share expertise, methodologies, and research data. The various research programs will investigate the effect of irradiation on a variety of human pathogens (e.g., *E. coli* O157:H7, *L. monocytogenes*, *Salmonella* spp., and *Shigella* spp.) associated with a number of whole and cut fruits, including apple, apricot, blueberry, cantaloupe, jackfruit, mango, peaches, pineapple, pomello, and watermelon. Additional research topics will include other vegetables, modified atmosphere packaging (MAP), sensory response, processing conditions, effect on shelf life and storage, etc. It is expected that the results of this multiyear research program will be used by food processors to design proper protocols for the irradiation treatment of fresh and minimally processed produce, as well as to improve the safety and quality of these fruits and vegetables.

This chapter will discuss the use of ionizing radiation in the processing of fruits and fruit products such as juices, ciders, and fruit pulp. While the focus is on fruits and fruit products, valuable insights may be obtained by an examination of the research conducted on irradiation of vegetable products, and these studies will be discussed where appropriate. The key technologies used for irradiation will be compared, and the current state and extent of commercial fruit irradiation will be summarized. Also discussed will be matters of quality control in irradiation processing of fruits, and ways that other treatments may be combined with irradiation in a "hurdle"-type synergistic control program, as well as the regulations and scientific data concerning packaging materials. In addition to a review of the relevant scientific information on radiation microbiology and physiology, this chapter will present summaries of the national and international regulations concerning which fruit commodities may be irradiated, the permitted purposes for irradiation, and the maximum doses allowed by key exporting and importing nations.

11.2 IONIZING RADIATION PHYSICS AND TECHNOLOGIES

The level of treatment received by a food product — the radiation dose — is defined as the quantity of energy absorbed during exposure. The international unit of treatment is gray (Gy). One gray represents one joule of energy absorbed per kilogram of irradiated product, the equivalent of 100 rad or 0.1 Mrad. The energy absorbed depends on the mass, bulk density, and thickness of the food. Each kind of food has to absorb a specific dose of radiation for the desired results to be achieved. An excessive dose may damage the food and make it unacceptable for consumption,

TABLE 11.1
Technologies for the Production of Ionizing Radiation

Factors	Electron Beam	X-Ray	Gamma
Source	Electrons are generated on an emission coil and accelerated to high energy, 5–10 MeV ^a . Extensive cooling equipment required	Created when high-energy electrons (up to 5 MeV) strike a metal plate (e.g., tungsten or tantalum alloys); typical conversion efficiency is 5–10%. Extensive cooling equipment required	Radioactive decay of cobalt-60 (2.5 MeV) or cesium-137 (0.51 MeV). Cooling equipment required
Mechanism	High-energy electrons cleave water molecules, creating oxygen and hydroxyl radicals that damage DNA membranes. Direct cleavage of DNA also occurs	High-energy photons stimulate atoms within target to release high-energy electrons that cleave water molecules into radicals. Direct cleavage of DNA also occurs	High-energy photons stimulate atoms within target to release high-energy electrons that cleave water molecules into radicals. Direct cleavage of DNA also occurs
Shielding required	During operation, > 2 m concrete or ~0.7 m steel/iron/lead. When source is powered off, no radiation is emitted	During operation, > 2 m concrete or ~0.7 m steel/iron/lead. When source is powered off, no radiation is emitted	> 5 m water or > 2 m concrete or ~0.7 m steel/iron/lead. Source cannot be turned off, shielding of source must be the default position
Speed ^b	Seconds	Seconds	Minutes (depending on source strength)
Penetrability ^c	6–8 cm, suitable for relatively thin or low-density products. Passes from multiple angles may be required	30–40 cm, suitable for all products	30–40 cm, suitable for all products

^a MeV = million electron volts.

^b Speed of dose delivery. The desired dose will vary depending on the target organism and commodity irradiated.

^c Penetrability in food products of average density approximating 1 g/cm³. This figure will vary for individual commodities due to localized variation in density associated with bone, voids, fibrous matter, etc.

while an inadequate dose will fail to achieve the desired effects. The qualitative terminology used to describe doses is not firmly established, but generally, doses may be characterized as low (< 3 kGy), medium (> 3 and < 10 kGy), or high (> 10 kGy).

The three types of ionizing radiation that are used for commercial food processing are gamma rays, e-beam, and x-rays. An overview of the advantages and disadvantages of each is presented in Table 11.1. Each of the technologies, whether high-energy photon or high-energy electron, induces ionization of molecules in the food target, leading to the generation of radicals, breakage of DNA, and other radiochemical effects. While these types of radiation are energetic enough to ionize atoms in the food being treated, it should be noted that they are not energetic enough to induce radioactivity in the target. This process is discussed in greater detail in the Section, “General Mode of Action.” Ozone and heat are generated during the irradiation process for each of the technologies; proper ventilation and temperature control are therefore especially important for the irradiation of fruits to avoid product degradation from these secondary effects [29].

11.2.1 GAMMA RAYS

Gamma rays are high-energy photons produced by the disintegration of radioactive isotopes. The isotopes of significance in food irradiation processing are cobalt-60 and, less commonly, cesium-

¹³⁷Gamma radiation is generated when cobalt-60 (half-life of 5.27 years) disintegrates. It first produces an unstable intermediate isotope of nickel-60 with emission of beta-radiation and then disintegrates further into stable nickel-60 with emission of two gamma rays of average energies of 1.17 and 1.33 MeV, respectively. Disintegration of cesium-137 (half-life of 30.17 years) to barium-137 produces gamma rays with an energy of 0.66 MeV.

The preferred radioisotope for food irradiation has historically been cobalt-60. Gamma rays from cobalt-60 are more energetic than those from cesium-137, but due to its much shorter half-life, cobalt-60 irradiators must be replenished with fresh radioisotope (recharged) much more frequently than cesium-137 irradiators. After 10.5 years of operation, a cobalt-60 irradiator will have approximately 25% of its initial strength, while a cesium-137 irradiator will have retained approximately 80% of its initial strength. Despite the need for more frequent recharging, the key factors that have made cobalt-60 the preferred radioisotope include the higher initial cost of cesium-137 and the water solubility of the CsCl form of cesium-137 used in irradiators, a point of significant environmental concern.

Gamma rays have excellent penetrability and are suitable for irradiating large food items such as pallet- or crate-sized packages of product. The shielding required for a gamma irradiator is roughly comparable to that required for an e-beam or x-ray irradiator (Table 11.1). However, the stigma attached to radioactive material, particularly in a food processing facility, as well as the always-on nature of the source, has led to only limited adoption of gamma sources for food processing. The time required for processing is dependent on the dose desired and the strength of the source. Longer processing times may necessitate some form of temperature control, particularly in temperature-sensitive products such as fruits.

11.2.2 ACCELERATED ELECTRON BEAM (E-BEAM)

E-beams are produced by electronic equipment that uses a linear accelerator or a cyclotron accelerator to impart a high velocity, and therefore a high kinetic energy, to a stream of electrons. The accelerators used in commercial irradiators intended to treat food produce a focused beam of electrons with an energy of up to 10 MeV. The processing dose is delivered as a pulse of electrons, and the full dose is delivered quickly, taking typically less than 5 sec. The electrons are aimed at the target with a cone-shaped guide. Higher doses may be delivered by repeated exposure. Despite the intensity of the beam, the short exposure time for product being irradiated generally prevents any significant rise in temperature during processing.

The penetrability of the electron beam is lower than that of gamma rays or x-rays (Table 11.1), so product must be packaged in relatively thin cartons treated from multiple sides or some combination of these. The increased handling (e.g., stacking and unstacking pallets of produce, multiple passes, etc.) may result in decreased throughput compared to gamma ray or x-ray processing, as well as increased opportunities for postprocess handling damage to the product. With regard to construction of new irradiation facilities, a significant commercial advantage of e-beam irradiators over gamma irradiators is the electronic nature of the radiation source, which enables it to be completely deactivated — i.e., when it is off, it is off. It is important to note, however, that an electron accelerator does not resemble a home microwave oven; as high-power, industrial-scale electronic devices, electron accelerators require specialized training and maintenance for their operation, and the shielding required for an operating e-beam unit is comparable to that required for a gamma irradiator.

11.2.3 X-RAYS

X-rays are high-energy photons and are the form of ionizing irradiation most familiar to the general public due to their widespread medical applications, although at much lower energies. The high-energy x-rays used in food irradiation are generated from high-energy electrons and thus require the same type of accelerator systems used in an e-beam irradiator. The beam of electrons is directed at a high-density metal target. The electrons are absorbed by the metal atoms and the high-energy

photons (x-rays) are emitted. The efficiency of this process is relatively low, typically 5 to 10%, depending on the specific alloy used in the metal target plate. Alloys of tungsten, tantalum, and various types of stainless steel have been used, and they vary in durability, cost, and efficiency.

The energy of the x-ray output is dependent on the energy of the originating e-beam and the conversion efficiency. The vast majority of the energy that is directed at the metal plate is converted into heat, making the proper design of cooling subsystems in x-ray irradiators critical. Also of concern are neutrons that are occasionally knocked from the high-density metal plate by the e-beam. At high energy levels, these neutrons can induce a low level of short-lived radioactivity in the target [36]. This neutron generation is less likely, although not impossible, with the lower-density foodstuffs targeted directly in e-beam irradiators.

Shielding for an x-ray irradiator is comparable to that required for the other two types of units. X-rays, like gamma rays, are high-energy photons, have a similar penetrability (Table 11.1), and can be used for bulk packages or higher-density foodstuffs. However, x-rays share with e-beam the commercial advantage of being produced electronically and are therefore able to be completely inactivated in a power-off state. The design of some commercial e-beam irradiators includes the ability to convert from e-beam to x-ray operation as needed by installation and removal of the metal target plate. While x-rays would seem to be an attractive middle ground between gamma and e-beam irradiators, the low energy conversion efficiency, the extensive heat build-up in the metal plate, and the low, but measurable, neutron scattering are drawbacks for commercial application.

11.2.3 GENERAL MODE OF ACTION

A stream of high-energy photons, either gamma rays or x-rays, can energize electrons within the atoms of the target. These electrons may leave the atom completely (ionization), or the energy of the electrons may rise to a higher level within the atom (excitation). Both processes may yield free radicals, i.e., atoms with unpaired electrons on their outer shell. The stream of high-energy electrons in an e-beam interacts with the atoms directly to create free radicals. These free radicals are very reactive because their unpaired electrons may pair up with the outer shell electrons of the atoms that make up cellular components. Because water makes up the bulk of mass of foods, particularly fresh fruits and vegetables, the water molecule is most frequently affected. The majority of the absorbed energy from the ionizing radiation treatment goes into the creation of hydrogen and hydroxyl radicals from water molecules [26]. The interaction of these free radicals with the organic molecules of the food is the main mode of action of ionizing radiation. Under conditions of limited free water, such as in dried or frozen products, radicals are produced with less efficiency and have reduced mobility. In these products, higher doses become necessary for microbial control [37, 38].

In most discussions of food irradiation, a recurrent question arises regarding the mode of action of high-energy photons (gamma or x-ray) vs. high-energy electrons (e-beam), and the possibility of differential antimicrobial efficacy or effect on product sensory attributes or physiology [39]. It is tempting to make a blanket statement that, based on the physics of energy transfer in the process of generating radical molecules, one would expect to see no appreciable difference in irradiated product based on the means of irradiation. However, direct-comparison experiments to verify this contention with fresh produce are lacking. Papaya treated with gamma radiation tended to be firmer than fruit similarly treated (0.4 or 0.6 kGy) with x-rays; significant, though small, differences in aroma, texture, and color were detectable to one set of triplicate sensory panels but not to another [40]. In the available direct-comparison studies of meats, high-energy photons and high-energy electrons tend to have similar, but not quite identical, effects. E-beam and gamma (1.5 or 3 kGy) were similarly effective at eliminating *Salmonella typhimurium* from refrigerated beef steaks, but e-beam was not as effective as gamma in the elimination of *Pseudomonas fluorescens* (a common spoilage bacterium found on meats and produce) [41]. Additional direct comparisons of the various irradiation methods are therefore warranted, especially studies that involve sensitive products such as fruits and vegetables.

11.3 CHEMICAL EFFECTS OF IONIZING IRRADIATION IN FOODS

Although irradiation shows considerable promise for extending shelf life and maintaining microbiological and sensory qualities of fruits, the literature is rich in contradictions about the results of irradiation of specific fruits. This leads to difficulty in standardizing the treatment. The success of the treatment depends on numerous factors. The most important ones are commodity and cultivar, dose of radiation, degree of maturity, physiological status of the fruits, temperature and atmosphere during and after treatment, pre- and postharvest practices, and susceptibility of the microorganisms to be controlled to radiation.

11.3.1 MACROMOLECULES

Nucleic acids, because of their large size, are the main targets of free radicals generated by gamma rays. They may be affected in various ways, leading to breaks in one or both strands of DNA or to intra- or extramolecular cross-linking. Polysaccharides, cellulose, pectins, and starches may be partly depolymerized. Proteins are relatively little affected, although reduction of disulfide bonds may lead to inactivation of active sites and conformational changes in enzymes.

11.3.2 SMALL MOLECULES

Sugars may be hydrolyzed or oxidized when subjected to gamma radiation. Free amino acids can be deaminated. The small molecules most affected are the polyunsaturated fatty acids. Free radicals react with polyunsaturated fatty acids, producing unstable hydroperoxides and a range of further degradation products. The effect of irradiation on vitamins has been studied extensively. Certain vitamins (A, B12, C, E, K, thiamine), particularly those with antioxidant activity, are degraded when irradiation is carried out in the presence of oxygen. Loss of vitamin C is commonly overestimated in the literature because ascorbic acid is oxidized to dehydroascorbic acid that is still active as vitamin C. Dehydroascorbic acid is, for the greater part, reduced back to ascorbic acid in living plant tissues. Only when dehydroascorbic acid is further oxidized to diketoglutaric acid is it irreversibly lost as vitamin. In general, small molecules, including vitamins, are little affected by low-level irradiation, and to a lesser extent, by thermal processing.

11.3.3 PROTECTION IN COMPLEX FOODS

Antioxidants added to solutions are known to protect sugars, vitamins, and proteins, as well as suspended bacteria from the effects of ionizing radiation [42, 43]. It may therefore be theorized that naturally occurring antioxidants may serve to protect plant tissue from radical molecules. However, the role of natural antioxidants in determining radiation tolerance of fruits and vegetables has not been addressed directly. Kader [15] and Dupont et al. [44] grouped fruits and vegetables according to their tolerance for irradiation. Products that are more radiation tolerant are, in some cases, also reported to be higher in antioxidant concentration, and lower radiation tolerance was similarly associated with low antioxidant concentration [45, 46]. However, methodological differences in how the produce was evaluated in the various studies prevent definitive conclusions. It is important to note that each commodity, and even each cultivar, has its own limits of tolerance. The dose to be applied should be preliminarily tested on a sample.

11.4 BIOLOGICAL EFFECTS OF IONIZING IRRADIATION IN FOODS

11.4.1 PRINCIPAL TARGETS

The principal targets of the radical molecules, with respect to biochemical function, are nucleic acids and membrane lipids. Alterations in DNA will affect gene expression and the biosynthesis

of various enzymes and will interfere with cell division. DNA is generally more sensitive to damage from radicals during active replication or transcription [47]. Bacterial plasmid DNA may also be damaged, disrupting and inactivating plasmid-encoded genes; pathogenic bacteria that carry plasmid-encoded virulence genes may therefore have a reduced virulence after irradiation [48].

Alterations in membrane lipids, particularly polyunsaturated lipids, lead to the perturbation of the membrane and to deleterious effects on various membrane functions — most important, permeability. Proteins are scarcely affected, and enzyme inactivation is rarely observed when irradiated *in situ*. In pure solution, peptide bonds and sulfhydryl groups, often involved in enzyme activity sites, may hydrolyze or oxidize into disulfide intra- or inter-peptide bridges. However, the activity of membrane-associated enzymes may be affected as a secondary effect of membrane lipid degradation.

11.4.2 SENSITIVITY OF ORGANISMS

The physiological effects of ionizing radiation on spoilage organisms and human pathogens have been reviewed by Monk et al. [49] and Diehl [26]. Fungi are, typically, more resistant to radiation than bacteria [49, 50]. D_{10} values, i.e., the amount of radiation necessary to effect a 90% (1-log) reduction, have been reported for yeasts and molds in the range of 1 to 3 kGy [51–53] as opposed to D_{10} of 0.3 to 0.7 kGy for pathogenic bacteria on produce and fruit juices [54–57]. Human pathogenic viruses are usually more resistant to radiation than bacteria or fungi. The doses required to achieve meaningful virus population reductions, as a rule, result in loss of sensorial quality of the produce [27, 30, 31, 49]. D_{10} for plant pathogenic fungi has been shown to range from 0.4 to 1.1 kGy [58]. Low dose irradiation has been shown to suppress some plant pathogenic fungi responsible for storage losses [16]. However, the relative resistance of fungi suggests that irradiation is best suited to control of bacterial pathogens, as opposed to viral or fungal pathogens.

A concern expressed by various authors [59] is the possibility of increased human or plant pathogen growth in storage following irradiation due to reduced interspecies competition. Although studies of the effect of low radiation doses on the microbial ecology of produce are lacking, studies of irradiated meats have tended to support the position that while irradiation of a product reduces the total population of bacteria on the sample, it does not lead to increased recolonization by pathogens [59, 60, 61]. However, Matches and Liston [62] found that salmonella grew more rapidly on irradiated vs. nonirradiated fish fillets. Carlin et al. [63] showed that the growth of *L. monocytogenes* inoculated onto previously disinfected leaves of endive (*Cichorium endiva*) was significantly enhanced. While this study [63] relied on chemical disinfection rather than irradiation, the result indicates that the interaction of native microflora and bacterial pathogens such as *L. monocytogenes* is an important subject for future study.

11.5 TECHNICAL ASPECTS OF FOOD IRRADIATION

11.5.1 DOSIMETRY

Measurement of the dose of radiation absorbed by the food (dosimetry) is the underlying key to effective use of irradiation. It is important to stress that the quality of the dosimetry will determine the quality of the irradiated product. Material near the outside of a carton or package tends to receive a somewhat higher dose than the product in the center, owing to its proximity to the radiation source. The ratio of the maximum amount absorbed (typically, near the perimeter) to the minimum absorbed (typically, near the center) is referred to as the max/min ratio. In an ideal, perfectly uniform treatment system, this value would be 1; in practicality, max/min ratios of 1.5 to 2 are more typically obtained. The max/min ratio is influenced by the bulk density of the product being treated, the penetrability of the radiation type, the strength of the source, and localized density variations within the product (voids, pits, etc.). A Monte Carlo simulation of a single apple's dose absorption profile suggests that even within a single piece of fruit, the max/min ratio may be in

excess of 2.4 due to the topographical complexity of apples [64]. A process that has a high max/min ratio may result in one part of a shipment receiving too high a dose (resulting in violation of regulations, sensory damage to product, etc.) while another part receives too low a dose (resulting in surviving insects or bacteria, inadequately delayed ripening or sprouting, etc.). Thus, design of effective irradiation protocols begins with a preliminary 3-D mapping of the doses absorbed by the product in each part of the package, when it is packaged in the type of carton and packing arrangement expected to be used commercially.

Dosimetry is important for establishing the desired dose for each commodity, for obtaining data required for approval of a petition by regulatory agencies, and for developing quality control procedures in the irradiation plant. Dosimetry is also used to determine the configuration of the irradiation field in the facility. McLaughlin et al. [65] have reviewed the technical aspects of dosimetry in detail. Although the absorbed dose can be calculated, experimental dosimetry is preferred. The energy absorbed is measured by placing dosimeters (radiation sensors) inside the lot being irradiated. In this way, the distribution of the energy absorbed and the minimum and maximum dose can be determined. The choice of dosimeter is influenced by several factors, including environment (humidity, temperature, and light) stability under the process conditions, dose range, and dose rate. It is therefore important to have a quality control procedure to test the dosimeters.

An example of a chromatographic dosimeter commercially available for routine measurement of absorbed dose is the Harwell YR (Didcot, U.K.). This is a radiation-sensitive polymethylmethacrylate-based product that darkens on exposure to ionizing radiation. Chromatographic dosimeters are read with a spectrophotometer, and the extent of color change is calibrated to absorbed dose. Electron paramagnetic resonance (EPR) is a sensitive dosimetry technique suitable for research purposes. The dosimeter consists of an alanine pellet that undergoes a measurable shift in paramagnetic spin state following exposure to ionizing radiation. With proper storage, the dosimeter pellet can hold the new electron spin state for months, providing an opportunity for repeated reference to the same set of processed dosimeters.

11.5.2 DETECTION OF PRIOR IRRADIATION

Although irradiation of various food products has been approved by various nations, detection methods are not required for clearance. Development of methods to detect irradiation and to determine the dose absorbed are, nevertheless, important for a number of reasons, e.g., differences in national legislation, control of international trade, quality control, and consumer reassurance. Regulatory authorities and processors are the two groups that are most interested in the development of such methods. Whenever attempts have been made to detect irradiation on an experimental basis, the task has proven most difficult.

The changes are not radiation specific. It is probably not possible to develop a detection method for foods in general [66]; however, it is possible to devise such methods for specific food groups. Detection of irradiation in food has been reviewed in detail by Raffi [67] and Delincee [66]. Detection of irradiation has been investigated for a large number of fruits including banana, mango, papaya, and strawberry [Figure 11.1; 68]. These methods were usually based on chemical, physical, and biological changes in fruits. Some methods studied are listed below.

After irradiation, carbohydrates may react to give acids and carbonyl compounds. Den Drijver et al. [69] detected glucosone (D-arabinohexos-2-ulose) in an irradiated sugar mixture solution mimicking the sugar composition of the mango, and not in the untreated controls. Studies with apples showed significant changes in starch and pectins [70, 71]. Detection of irradiation based on DNA analysis is promising and widely applicable; however, more research is needed to obtain results that are both specific and reliable [68]. Inhibition of seed germination (half-embryo test) has been used as an indication of exposure to radiation for citrus fruits [72]. It should be noted, however, that alfalfa seeds treated with 5 kGy did not show a significant change in germination rate, suggesting that caution should be exercised with this method of detection [73].

Apple	Citrus	Lemon	Passion fruit	Red currant
Avocado	Fig	Mango	Peach	Strawberry
Banana	Gooseberry	Melon	Pear	Tomato
Blackberry	Grape	Mulberry	Plum	Watermelon
Black currant	Grapefruit	Orange	Prune	
Cherry	Kiwi	Papaya	Raspberry	

FIGURE 11.1 Fruits investigated for detection of radiation treatment. (Adapted from IAEA, Analytical Detection Methods for Irradiated Foods, IAEA-TECDOC-587, Vienna., 1991.)

EPR was shown to be reliable with strawberry achenes exposed to a dose of 1 kGy or more [74] and can be used with blueberry, fig, raspberry, and red currant. EPR can also detect prior irradiation of alfalfa seeds (D.W. Thayer, personal communication). The test may not be reliable with all fruits, as shown with plums [75] and with citrus and grapes [68]. Detection of radiation-induced hydrocarbons by gas chromatography or mass spectrometry has been successfully employed in irradiated (1 kGy) perilla (*Perilla frutescens*) seeds [76]. Rahman et al. [77] showed that the irradiation dose used for quarantine treatment induced a significant decrease of the size of the supraesophageal ganglion of insects.

Measurement of luminescence is the first detection method used routinely for inspection purposes. The method, used in Germany, is based on the determination of light emitted by oxidizing substances formed and trapped in irradiated tissue. It can be used with produce containing solid inclusions, such as achenes and adhering minerals. Chemiluminescence, resulting from oxidation reactions after irradiation, was used to detect irradiation of spices, dried herbs, and fruits [78, 79]. Thermoluminescence is released after heating of irradiated products. It is more reliable than chemiluminescence since it is less influenced by interferences from test conditions [68]. This approach gave good results with strawberries [79]. Thermoluminescence has shown promise in detecting irradiation treatment of spices [81] and potatoes [82].

Two methods that showed some potential with meat were tested on fruits with limited success. Changes induced to the microflora by irradiation were too influenced by pre- and postharvest conditions to be reliable. Determination of *o*-tyrosine in proteins was attempted with strawberries but showed high background values and artifacts [83, 84]. Changes in the content of phenolic compounds [85] may not be specific to radiation because such changes are common under stress conditions.

Presently, the more promising irradiation detection methods for fruits seem to be EPR and thermoluminescence. A particular challenge is the low dose of irradiation, 1 kGy or less, with which fruits are treated. Further investigation, standardization, and development of instrumentation will eventually lead to practical inspection methods. It should be noted that *ex post facto* dosimetry is difficult; while these and other detection methods may be able to indicate that a given product was irradiated, it is unlikely to determine when the product was so treated, or when the dose was delivered [26].

11.6 IRRADIATION FACILITIES

Although e-beam, x-ray, and gamma radiation have essentially identical effects on food, irradiation facilities differ greatly with respect to design and physical arrangement. The vast majority of gamma-based facilities use cobalt-60 as the radionuclide source. The majority of new irradiation

Ionizing Radiation Processing of Fruits and Fruit Products

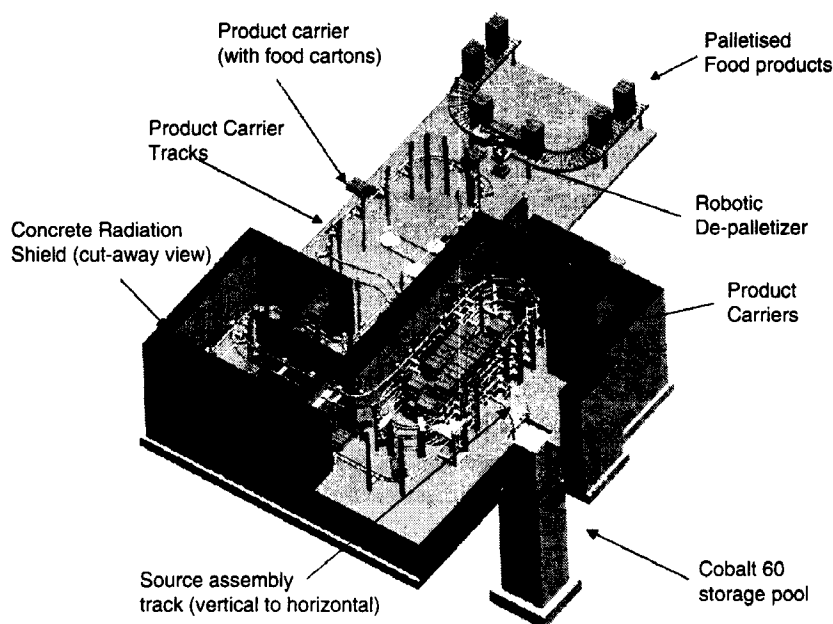


FIGURE 11.2 Semiautomated cobalt-60 irradiator specially designed for processing food products. (From Nordion International Inc., Kanata, Ontario. With permission.)

facilities under construction in the U.S. are e-beam or dual-use e-beam and x-ray facilities. Several new e-beam facilities have been built in the U.S. in the last decade for irradiation of meat and poultry products, shell eggs, and Hawaiian fruits destined for the U.S. mainland, such as papaya. In 2001, the U.S. Postal Service contracted to purchase eight e-beam systems for irradiation of mail in conjunction with antiterrorism efforts, with options to purchase as many as 12 additional facilities. Thus, in terms of market share and processing capacity, e-beam technology may benefit from increased familiarity with the technology and the economies of scale that come with mass production of the equipment.

Figure 11.2 depicts a typical cobalt-60 automatic carrier irradiator specifically designed for processing food products (Nordion International Inc., Kanata, Ontario). This unit is suitable for processing large volumes (up to 8 million ft³ annual throughput) in continuous operation. Produce is loaded into boxes on a conveyor. The boxes move around the cobalt-60 source within a thick concrete enclosure shielding the environment from the gamma rays. The source is lowered into a pool of water when not in use. The dose applied is controlled by regulating the speed of the conveyor.

In 2001, a joint venture between an American manufacturer of e-beam/x-ray equipment (Sure-Beam, San Diego, CA) and a Brazilian irradiation processing corporation (Tech Ion, Manaus, Brazil) was announced [86]. This joint venture is expected to facilitate irradiation of fruit and vegetables produced in Brazil, with the stated goals of reducing postharvest losses in storage and transshipment, and improving disinfestation. A gamma-based irradiation facility is currently in operation in Manaus, Brazil. Fruits irradiated at this facility are destined primarily for the large population centers of Sao Paulo and Rio de Janeiro in the southeast [87].

As part of a significant effort to promote and facilitate food irradiation, the government of Brazil recently established the Food Irradiation Center of Excellence in Rio de Janeiro [86]. Near the city of Rio de Janeiro, a large-scale irradiation facility is under construction, which is designed to incorporate e-beam, x-ray, and gamma-ray processing capacity at a single site [35]. This facility is expected to be operational in 2002 with an accelerator of 15 kW power at 10 MeV, with an expected capacity of 120,000 t/year. The processing capacity of the x-ray processing unit is expected

to be 80,000 t/year. The cobalt-60 source strength is planned to be 600,000 Ci with a capacity of 120,000 t/year. This facility will process fruits and vegetables intended for domestic consumption, as well as for export.

An x-ray facility near Hilo, HI, is currently irradiating fresh fruits intended for shipment to the mainland U.S., with particular focus on the New York, Washington, Oregon, California, and Minnesota markets. The fruits treated include papaya, rambutan, lychee, atemoya, longan, and star fruit [88]. The fruits are treated with 0.25 to 0.40 kGy for disinfestation purposes; an extended shelf life is an additional benefit to the treatment. The projected production capacity of this facility is 12,500 t/year.

11.7 CONTROLLING REGULATIONS: COMMODITIES, DOSE LIMITS, AND PURPOSES

Each nation issues its own regulations concerning the commodities that it allows to be irradiated, the maximum (and in some cases, the minimum) dose to be applied, and the purposes for which a product may be irradiated. In many cases, the intended purpose of the irradiation treatment will determine the maximum dose that can be legally applied. Regulations concerning the approval and inspection of food irradiation plants are also nation specific. It is expected, however, that as food processing operations, irradiation facilities will be held to the highest standards of cleanliness and safety. The regulations governing products, dose limits, purposes, etc., can range from very restrictive to very permissive. The International Consultative Group on Food Irradiation (ICGFI) is a group of experts from 46 nations organized under a joint FAO/IAEA/WHO collaboration to review and disseminate scientific information regarding food irradiation. ICGFI maintains an online listing of the regulatory limits governing food irradiation for a variety of developed and developing nations [89].

This section will provide an overview of some key nations with regard to regulations dealing with fruits and vegetables. With changing consumer attitudes toward irradiated foods, the implementation of irradiation on food is increasing worldwide. The controlling regulations in many nations are currently being reviewed and revised, and the current state of affairs may therefore differ somewhat from the information presented herein.

U.S. regulations permit the irradiation of dry or dehydrated vegetable-derived spices, seasonings, and flavorings, as well as coloring agents (e.g., paprika) to doses of up to 30 kGy [90]. Approved sources are gamma rays from cobalt-60 or cesium-137; accelerated electrons from a machine source, not exceeding 10 MeV (to eliminate the risk of inducing radioactivity); or x-rays from a machine source, not exceeding 5 MeV. The regulatory limit for fresh fruits and vegetables is 1 kGy, limited to the specific uses of disinfestation and inhibition of produce growth and maturation [90]. For comparison, irradiation to control food-borne pathogens is permitted at higher doses for shell eggs (3 kGy), seeds used for growing sprouts (8 kGy), fresh and frozen poultry (3 kGy), fresh meats (4.5 kGy), and frozen meats (7 kGy). Irradiated foods must bear the internationally recognized radura logo (Figure 11.3) along with the statement "treated with (or by) irradiation." In 1999, a coalition of U.S. food processors petitioned the U.S. FDA to amend U.S. regulations to allow doses of up to 4.5 kGy for a wide variety of refrigerated and ready-to-eat meat and vegetable products, including juices, and doses up to 10 kGy for frozen meat, vegetable, and juice products. Elimination of human pathogens is the primary goal of these requested dose limits; the potential for extension of shelf life is regarded as a secondary goal [91].

Many European nations (e.g., Austria, Germany, Ireland) have approved only dry herbs, spices, and vegetable seasonings for irradiation, with a maximum dose of 10 kGy. Canada has approved for herbs, spices, and seasonings (max 10 kGy), and allows irradiation of onions and potatoes for sprout suppression (max 0.15 kGy). Mexico will allow any type of fresh fruit to be treated with up to 1 kGy for quarantine treatment or to delay ripening, and up to 2.5 kGy to extend shelf life.



FIGURE 11.3 The *Radura* — the internationally recognized logo identifying foods that have been irradiated. AU: Is this correct?

Mexican dried fruit may be treated with 1 kGy for disinfestation but with up to 10 kGy for microbial control. The U.K. allows fresh or dried fruit to be treated for disinfestation (max 2 kGy). India has approved irradiation disinfestation of dates and mangoes (max 0.75 kGy). Japan has approved treatment of a single product, potato, for sprout inhibition (max 0.15 kGy). Australia allows irradiation of herbs and spices for disinfestation (max 6 kGy) or microbial control (max 30 kGy).

Brazil possesses the most open regulations concerning food irradiation. Prior to 2001, a specified list of fruit commodities were permitted for irradiation [87]. This list included avocado, banana, orange, lemon, and other fruits important for domestic consumption and the export market. The maximum dose permitted was 1 kGy, although the treatment could be applied for a variety of purposes, including disinfestation, delay of ripening, shelf-life extension, or (in combination with a heat treatment) control of microbial load. A joint FAO/IAEA/WHO study [92] examined the wholesomeness of food irradiated above 10 kGy. In that study, it was concluded that food may be safely irradiated to any dose sufficient to achieve the desired physiological or microbial outcome, without appreciable loss of nutritional adequacy. Based on this recommendation, the Brazilian government dramatically revised its regulations governing food irradiation [89]. As of 2001, any food product in Brazil, such as fruit, vegetable, meat, poultry, and fish, etc., may be irradiated to any dose for any purpose. The only limit on the dose applied is based on the product's physiological tolerance for irradiation from a quality or marketability standpoint.

Brazil's "any product, any dose, any purpose" stance, although based on the scientific recommendations of respected international authorities, is markedly more open than most other nations. Brazilian products that are intended for export must still meet the regulatory requirements of the importing nations, and are therefore, as a matter of practice, subject to dose limits and purpose restrictions. Although efforts are underway to encourage Brazil's trading partners to lift restrictions on irradiated foods (J.F.B. Medeiros, personal communication), dramatic changes to the regulations of importing nations are not expected in the near future.

11.8 APPLICATIONS OF IRRADIATION TO FRUIT

Fresh produce may be irradiated for a number of purposes, including inhibition of sprouting in the case of some vegetables, and, of more significance for irradiated fruit, delay of ripening, insect

disinfestation, and reduction of microbial load. Of these, the most important effect of irradiation in food processing, whether meats and poultry or fresh fruits and vegetables, is reduction of microbial load. While this has traditionally meant elimination of produce spoilage pathogens so as to extend shelf life [93, 94], the elimination of human pathogens is also of increasing importance [16, 17, 25]. Refrigeration and modified-atmosphere packaging are currently the primary means of preserving fresh produce [20]. Fresh produce is generally more sensitive to ionizing radiation than meat or poultry products. Unlike meat, fresh produce is living tissue that respire, maintains water relations with its environment, and, in many cases, may continue synthesis of secondary metabolites [95]. Irradiation can alter these processes, leading to changes in firmness, aroma, color, or taste [25, 30]. Plant physiology and surface-associated microbial ecology can be altered by irradiation; these effects are seen most clearly during subsequent refrigerated handling and storage [27, 82, 96].

In general, ionizing radiation causes less alteration to fresh produce than other preservation techniques with equivalent benefits, e.g., thermal treatment. However, irradiation does not replace these other methods; it supplements them. Irradiated fresh produce, like all irradiated foods, remain subject to the basic rules of good manufacturing practice for preservation of quality and food safety [2].

11.8.1 DELAY OF RIPENING

Ripening of fruits can be delayed by doses of 0.2 to 0.5 kGy. The underlying mechanism of the effect is not well understood. It involves interference with biochemical processes that are part of ripening and senescence [97]. Ethylene production by Gala apples was reduced by gamma irradiation (0.44 kGy) [98]. Most studies on fruit irradiation have been carried out with gamma rays. E-beams do not penetrate enough to act on metabolism and increase shelf life of certain fruits, e.g., avocado [99].

Tolerance varies with degree of maturity, as discussed by many authors. Delay in ripening of climacteric fruits requires that the treatment be applied before the onset of the climacteric increase in ethylene production [29, 97]. The need to treat at an early stage of maturity brings about a loss of quality, and may cause abnormal ripening and uneven coloring; however, in the case of tomato, very early treatment may accelerate ripening due to ethylene production in response to wounding [100].

11.8.2 DISINFESTATION

Disinfestation, the control of arthropod pests, can be achieved by doses up to 3 kGy. Sterilization to prevent reproduction of the insects during or after storage can be achieved by doses of 0.03 to 0.2 kGy. Doses sufficient to kill insects outright are in the range of 1 to 3 kGy. This application of irradiation is among the most promising since fumigation with chemicals like ethylene dibromide (prevented in the U.S. since 1984) is increasingly questioned. Irradiation is probably the most effective quarantine treatment for control of fruit flies and other insects in a number of commodities, including mango and papaya. Other quarantine treatments exist but are not easily applicable to all fruits. Cold treatment requires several days and is therefore not useful for fruits with short shelf lives. Heat treatment occasionally causes physiological disorders and discoloration.

11.8.3 REDUCTION OF MICROBIAL LOAD

The efficacy of ionizing radiation to reduce the microbial load is dependent on a number of factors. Plant fruits and stems typically support 10^3 to 10^6 colony-forming units (cfu) per gram of plant tissue [20, 101]. These organisms come from the environment in which the plants are grown, including the soil, water, air, manure, and compost, as well as from postharvest handling, processing, and shipping [24]. It is increasingly recognized that bacteria may become internalized in fresh produce and survive for days or weeks, reducing the efficacy of traditional, surface-oriented antimicrobial measures such as chemical rinses and washes [33, 102–105].

The vast majority of plant-associated microorganisms are nonpathogenic bacteria. These bacteria often form biofilms that can further reduce the efficacy of antimicrobial measures [106–108]. Human pathogens such as *E. coli* and *Salmonella* have been observed to form durable biofilms on industrial surfaces [109, 110]. The extent to which enteric bacteria, as well as the psychrotrophs *L. monocytogenes* and *Yersinia enterocolitica*, participate in preexisting phytoplankton biofilms formed by nonpathogenic bacteria is not known [108, 111]. Ionizing radiation can penetrate sheltered areas of fruits, and, in the case of gamma and x-ray, penetrate internal structures to inactivate bacteria. As has been discussed, however, the radiation types differ in penetrability, most significantly in higher-density products. In a study of sliced cantaloupe inoculated with human pathogens (10^6 cfu/g), after radiation treatment with 3 kGy from a 5-MeV e-beam, Draughon et al. [112] were able to recover salmonella and listeria, but not staphylococcus. The authors concluded that for the 1-cm thick slices examined, an e-beam dose of greater than 3 kGy would be necessary to completely eliminate salmonella or listeria.

Vegetables have been the object of numerous studies of the elimination and recovery of human pathogenic bacteria following irradiation. Total aerobic plate count and inoculated *L. monocytogenes* were reduced by ~4 logs on precut bell pepper following a dose of 1 kGy. The pathogen regrew to initial levels within 4 d on peppers subsequently stored at 15 or 10°C, but remained low on peppers stored at 5°C [113]. The authors concluded that irradiation followed by refrigeration effectively suppressed pathogen growth throughout the useful shelf life of the produce. *E. coli* and *L. monocytogenes* were effectively eliminated (> 5 logs) from diced celery by 1 kGy [28]. Also in that study, total aerobic counts were determined, following irradiation or one of three conventional treatments (i.e., acidification, blanching, or chlorination). Acidification reduced initial counts to a degree comparable to that of 1 kGy, but by the end of the storage period (22 d), the aerobic population following conventional treatments had regrown to equal (acidification) or exceed (blanching and chlorination) the untreated controls, while that of irradiated (0.5 and 1 kGy) celery remained significantly lower throughout the study.

The D_{10} value for human or plant pathogenic bacteria can range from 0.3 to 0.7 kGy [54–57]. Achieving a 5-log reduction, then, would require between 1.5 kGy and 3.5 kGy. Fresh produce will generally tolerate doses of 2 kGy, although variation exists. This suggests, therefore, that irradiation will most likely play a role as one of several options in preserving fruit quality and microbial safety. These other options might include mild thermal treatment, modified atmosphere packaging, ozone treatment, among other processes. A complete discussion of combination treatments is presented later in this chapter.

11.9 RESPONSE OF FRUIT TISSUE TO RADIATION TREATMENT

11.9.1 WOUND RESPONSE

Plant tissues are relatively sensitive to ionizing radiation and show a typical wound response, characterized by a transient increase in respiration and ethylene production, even at low doses. Such a wound response was shown after irradiation of strawberries [114]. The rate of respiration increased linearly with increasing dose of gamma irradiation. The effect was transient and the rate of respiration decreased back to preirradiation levels within 24 h for the lowest dose, 0.3 kGy, but slower with an increasing dose. The low dose of 0.3 kGy was insufficient to control mold development, as indicated by the progressive increase in rate of respiration in control and 0.3 kGy irradiated fruits. This progressive mold development could be prevented by treatment with a fungicide and by irradiation at 1 kGy. Ethylene production also increased after irradiation, but in contrast with respiration, it reached a maximum at 1 kGy. The latter observation suggests that irradiation beyond 1 kGy caused membrane damage since ethylene production is membrane associated. Similar results were obtained by Larrigaudiere et al. [115] with cherry tomatoes. Other typical wound (and stress) responses are (1) stimulation of phenol biosynthesis, particularly stim-

ulation of the first enzyme in the pathway phenylalanine-ammonia lyase (PAL) [116] that predisposes the tissue to enzymatic browning, and (2) an increase in enzymatic defenses against free radicals scavenged by peroxidase, catalase, and superoxide dismutase [117].

Notwithstanding the information presented herein and the extensive body of research on the subject of irradiation of fresh horticultural products (by 1983, more than 1150 publications had been compiled [118]), not enough is known about the reaction of these live tissues to treatment. Of particular interest is the interaction of irradiation with commonly used agricultural practices such as application of hormone or hormone-like chemicals.

11.9.2 DELAY OF RIPENING

The mechanisms whereby irradiation delays ripening in several fruits are not fully understood at the present time. The effect is most associated with climacteric fruits, i.e., fruits that show a transient increase in their rate of respiration and ethylene production at the onset of ripening. The majority of fruits of tropical origin are climacteric, and they have, therefore, a short shelf life. They are also sensitive to low temperature, which is the main preservation method for produce. The biochemical basis for the delay in ripening is discussed by Thomas [119]. Irradiation was shown to influence the respiratory pattern of the fruits [97] and to cause a shift from glycolysis toward pentose phosphate shunt in bananas and toward the glyoxylate cycle in bananas and mangos [119]. Dubery and coworkers [97] showed that irradiation did not truly delay ripening of mango, but that it interfered with some biochemical processes, including respiration, involved in senescence. Gamma irradiation promoted respiration in Gala apples that had been treated with an ethylene action inhibitor (1-methylcyclopropene) and subsequently stored at 20°C for 3 weeks [120].

An important mechanism whereby the treatment slows down the ripening in climacteric fruits is not fully understood but seems to be the reduction of ethylene biosynthesis and of sensitivity of the tissue to ethylene, probably related to alterations in membrane physical characteristics [29, 100, 121]. The inhibition of ripening of irradiated Bartlett pears was not reversed by subsequent exposure to ethylene, supporting the suggestion of inhibited ethylene action in the fruit tissue [122]. The production of ethylene and a variety of volatile esters and alcohols was inhibited in irradiated Gala apples by doses as low as 0.44 kGy [98].

11.9.3 POSTHARVEST DISEASE AND STORAGE LOSSES

According to Dubery et al. [97], the physiological status of the fruits at the time of irradiation is probably the most important factor influencing the response of the tissues. This status is, in turn, influenced by the preirradiation history of the fruits, e.g., mechanical injury, season, and humidity at the time of harvest. The response of each individual batch of fruits is therefore difficult to predict. One important element of success is rapid precooling of the produce and irradiation at the lowest temperature tolerable by the fruits.

The ripening inhibition in fruits that have been treated with low doses of ionizing radiation has a notable side effect with regard to plant pathogens. A dose of radiation that is too low to control fungal development directly may, still, indirectly result in reduced fungal disease. Fruits progressively lose their resistance to plant pathogens with ripening. Fruits that have had their ripening delayed retain a higher level of host resistance to fungal plant pathogens. As the spoilage fungi are unable to establish themselves, secondary bacterial plant pathogens such as *Erwinia* spp. are also less able to become established on the fruit, and overall microbial development will be delayed as an added benefit. As with all applications of irradiation to food products, the treatment functions best as a terminal processing step in a system where postirradiation recontamination is avoided. Also, to maximize the efficacy of the process, preharvest cultural practices should reduce, as much as possible, the initial microbial load on the fruit surface. It is preferable, therefore, that the fruits be packaged before radiation treatment. A brief discussion of the effects of irradiation on packaging materials is presented later in this chapter.

11.9.4 PHYSIOLOGICAL DISORDERS

When exposed to doses beyond their limits of tolerance, fruits suffer physiological disorders that cause undesirable symptoms, mainly tissue softening and enzymatic browning [15, 123]. Tissue softening is caused by partial depolymerization of cell wall polysaccharides, mainly cellulose and pectins, and by damage to cell membranes, which results in loss of intracellular water [30, 124, 125]. Enzymatic browning is an indication of decompartmentation due to damage to membranes, bringing phenolic substrates in contact with polyphenoloxidases [121]. A calcium dip pretreatment prevented loss of firmness and texture in irradiated diced tomatoes [126]. Also, as alteration of the physical characteristics of cell membranes results from oxidative attack on polyunsaturated fatty acids of membrane lipids by oxygen-free radicals, irradiating in an atmosphere with reduced oxygen contact can reduce these effects. However, under these conditions, efficiency of the treatment is also reduced. A high carbon dioxide atmosphere was shown to protect tissues from radiation-induced loss of membrane proteins [127].

11.10 QUALITY OF IRRADIATED FRUITS

11.10.1 TEXTURE, COLOR, AND SWEETNESS

Broad statements regarding the effect of ionizing radiation on fruit quality are difficult to make. It is generally true that irradiation tends to make fruits softer and sweeter, primarily through hydrolysis of pectins and release of sugars following depolymerization of carbohydrate polymers [30, 124]. Softer fruit are more susceptible to damage during handling and shipping. This fact, in addition to the desire to reduce the possibility of postirradiation recontamination, reinforces the role of irradiation as a terminal, postpackaging process step.

Strawberries softened following a dose of 1 or 2 kGy, and chemical analysis showed an association with an increase in water-soluble pectin and a decrease in oxalate-soluble pectin [30]. Irradiation enhanced the sweetness of strawberries by reducing titratable acidity in comparison with the unirradiated controls [51]. Titratable acidity was similarly decreased in stored apples following irradiation [120]. Gibberellic acid-treated grapefruit tolerated 0.3 kGy with little loss of quality of the fruit, pulp, or juice; however, pitting of the skin, softening of the fruit, and loss of juice quality rose to unacceptable levels following treatment with 0.6 kGy [128]. Firmness of papaya, rambutan, and Kau orange declined following x-ray treatment (0.75 kGy) [129].

Two blueberry cultivars, i.e., Brightwell and Tifblue, did not differ in their response to gamma radiation, with no negative impact after 0.5 kGy; both cultivars showed softening and loss of quality following 1 kGy [130]. The flavor and texture of Sharpblue blueberries were considered acceptable following electron beam irradiation (1 kGy) [131], but the flavor and texture of Climax blueberries similarly irradiated (1 kGy) [132] declined significantly.

Color, pH, postharvest decay rate, and other agronomic factors related to the irradiated blueberries were reportedly not affected by dose level in these studies. However, climacteric fruits that have been irradiated when mature, but before the fruits have ripened, may not ripen normally and may develop uneven coloring and skin discoloration [15]. As in the case of grapefruit, effects on skin color and blemishes can be dose dependent [128]. Fresh-cut cantaloupe melons were slightly bleached, softened, and off-flavored following 3 kGy, but these sensory attributes were unaffected by 1 kGy [133].

11.10.2 FLAVOR AND AROMA

Radiation-induced alteration of flavor and aroma is variable, and is influenced by the product tested, dose, fruit maturity, cultivar, storage conditions, and other agronomic production factors. With regard to taste, a distinction is made here between sweetness, which results from the balance of sugar content and acidity, and flavor resulting from the complex combinations of volatile, low

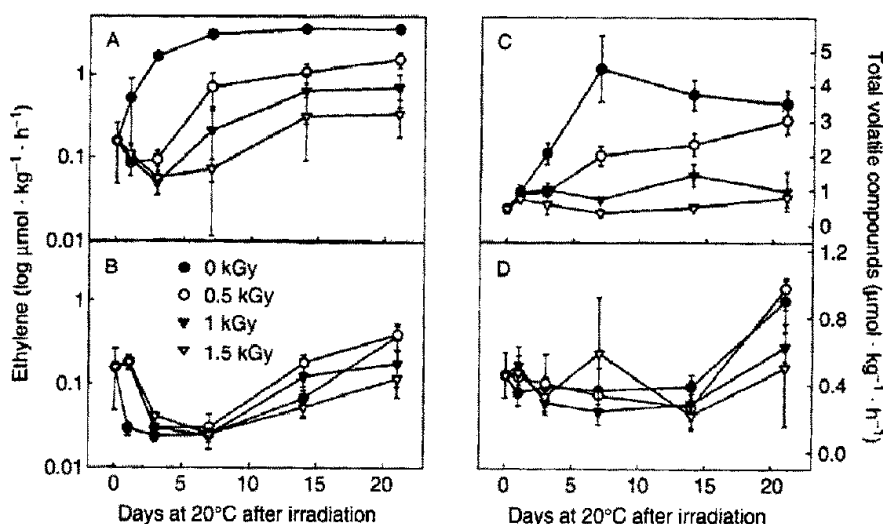


FIGURE 11.4 Production of ethylene (A, B) and total volatile compounds (C, D) of Gala apples during ripening. The fruit treated with (B, D) and without (A, C) $0.5 \mu\text{M}$ MCP were exposed to 0, 0.44, 0.88, and 1.32 kGy gamma irradiation and kept at 20°C. Vertical bars represent standard deviation. (From Fan, X. and Thayer, D.W., Quality of irradiated alfalfa sprouts, *J. Food Prot.*, 64(10), 1574–1578, 2001. With permission.)

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molecular weight compounds. This aspect of fruit quality may more accurately be referred to as the flavor–aroma complex as the two aspects are inseparably linked in fresh fruit. Modifications of taste had been reported by Kader [15] in the case of decreased acidity of strawberries and astringency in persimmons. The odor, color, and overall appearance of irradiated mango proved as acceptable to a sensory panel as nonirradiated controls immediately after treatment; the treated fruit remained acceptable after 50 d of refrigerated storage, while nonirradiated controls became unacceptable after less than 30 d [58]. Aroma and flavor of papaya, rambutan, and Kau orange were generally more intense following x-ray treatment (0.75 kGy) [129], although changes in other sensory characteristics were product specific.

Alteration of aroma is to be expected because many fruit aroma volatiles originate from the breakdown of polyunsaturated fatty acids, which are among the main targets of irradiation-generated free radicals. Fan et al. [98] examined the ability of Gala apples to generate volatile compounds after having been treated with gamma radiation, 1-methylcyclopropene (MCP), or both. MCP is an ethylene action inhibitor, and is used agronomically to delay ripening. Figure 11.4 shows the concentrations of ethylene and total volatile compounds generated by the apples in this study. In non-MCP-treated fruit, irradiation inhibited production of most esters and some alcohols in a dose-dependent response. In MCP-treated fruit, irradiation had little difference in ester and alcohol production over nonirradiated fruit. The authors conclude that MCP and irradiation result in comparable inhibition of volatiles in the apples.

11.10.3 NUTRITIONAL QUALITY

This aspect of food irradiation has been thoroughly investigated [15, 16]. Generally, low doses do not bring about significant losses of vitamins. The natural antioxidants, vitamins A, C, and E, are relatively labile, as are thiamine and vitamin B12. Irradiation is known to oxidize a portion of the total ascorbic acid (vitamin C) to the dehydro form [134]. While both the forms of this vitamin are biologically active, suggesting minimal nutritional impact [135], it is uncommon to see values for the dehydro form co-reported in analyses of ascorbic acid concentration. The impact of irradiation on vitamin C therefore tends to be overstated. Irradiated alfalfa sprouts showed (1) no

significant decline in total ascorbic acid concentration and (2) increased antioxidant power compared to nonirradiated controls [136].

11.10.4 INFLUENCE OF VARIETY/CULTIVAR

An important factor in the irradiation of fruits is consideration of variety- or cultivar-specific responses. Reviews of produce radiation sensitivity often combine different varieties under a single heading, e.g., “leafy vegetables” or “grape” [15, 44], a practice that does not recognize evidence of varietal differences. Plant variety or cultivar can influence the sensory response to irradiation, as well as the sensitivity of associated bacteria. Apple varieties show different degrees of internal browning and changes in firmness [120]. Blueberry varieties have cultivar-specific sensitivity, resulting in variable fruit quality [130–132]. In examining studies of irradiated vegetables, two potato varieties showed differences in the postirradiation (0.20 kGy) viscosity of their starches, whether isolated from irradiated tubers or irradiated *in vitro* [82]. Differences were seen in the respiration rates, loss of firmness, and in sensitivities associated with aerobic bacteria on iceberg lettuce [137] vs. romaine lettuce [96]. In a direct-comparison study, the radiation sensitivity of *E. coli* O157:H7 varied significantly when surface inoculated on iceberg or boston lettuce vs. green leaf or red leaf lettuce (B.A. Niemira, unpublished data).

11.10.5 QUALITY CONTROL

Because of the possible effects of irradiation on fruit quality at higher doses, it is essential to develop an extensive postirradiation quality control procedure. Basic quality criteria should be measured immediately after treatment and also after a few days of postirradiation storage for delayed effects. Surface color changes and internal discoloration are good indicators of injury. Respiratory rates, ethylene production, ripening, and senescence parameters, e.g., texture and color, give indications on the physiological effects of the treatment. Nutritional effects can be measured through determination of sugars, organic acids, pH, critical vitamins, or carotenoids. Enzyme assays, e.g., as related to enzymatic browning, may monitor biochemical effects. A rigorous sensory evaluation is an important element of treatment optimization and routine quality testing.

11.11 IRRADIATION OF FRUIT JUICES AND PULPS

The majority of fruit juices sold in the U.S. receive a conventional heat pasteurization treatment. The pasteurization process results in the loss of essential oils and other juice components, changing the flavor of the resulting juice. Fresh (i.e., nonthermally pasteurized) juices are valued for premium flavor and aroma; however, these products have also been responsible for outbreaks of salmonellosis, enterohemorrhagic *E. coli* infection, and hemolytic uremic syndrome [138]. Several strains of *E. coli* O157:H7 were found to survive in fruit pulps during extended refrigerated storage (up to 30 d in the case of grape and up to 20 d in the case of passion fruit pulp [139]). The U.S. FDA has implemented a policy [140] requiring 5-log reductions in human pathogen load in fresh juices, with these regulations to be in full effect for all processors by 2004. A variety of nonthermal means of reducing the microbial load of fresh juices were recently discussed [141]. These include pulsed electric fields, minimal thermal processing, high-pressure processing, and ultraviolet radiation. Regulatory approval for the use of ionizing radiation is currently being sought [91].

11.11.1 MICROBIOLOGY OF IRRADIATED JUICES AND PULPS

Preservation and shelf-life extension have been the historical focus of research on irradiation of juices. The key organisms of interest were yeasts and molds, which tend to have a higher D_{10} value than bacterial pathogens [49]. Juices and pulps, because of their high water content, represent an area that should provide the maximum opportunity for generation and mobility of radical molecules.

However, the same high water content means that the bulk density of these products is higher than what might be seen in packaged fruits. This must be taken into consideration with regard to the penetrability and max/min ratios attained during the irradiation process.

Phytopathogenic fungi were evaluated in irradiated mango pulp, with D_{10} values ranging from 0.39 to 1.11 kGy [58]. A complete discussion of this study is presented later in this chapter (see "Mango"). Niemira [56] reported a D_{10} value for *Salmonella enteritidis* of 0.35 kGy when irradiated in reconstituted orange juice. Buchanan et al. [54] showed that irradiation (1 kGy) effectively inactivated *E. coli* O157:H7 in inoculated commercial apple juices, with D_{10} values of 0.12, 0.16 and 0.21 kGy for the three isolates tested. Niemira et al. [57] found that the resistance of four salmonella isolates irradiated in orange juice also varied, with D_{10} values ranging from 0.35 kGy to 0.71 kGy. For the most resistant isolate (*Salmonella anatum*), 3.5 kGy was indicated as the dose required to achieve a 5-log reduction. Similarly, *Salmonella enterica* serotype Hartford was reduced to undetectable levels in irradiated (3 kGy) orange juice [142]. In that study, the pathogen was able to be recovered through selective enrichment.

The antimicrobial efficacy of irradiation is influenced by several factors. Native variation in pathogen resistance among different isolates has been noted [54, 57]. Buchanan et al. [54] showed that the radiation sensitivity of three strains of *E. coli* O157:H7 irradiated in apple juice was reduced by 54 to 67% by previous growth of the cultures in acid environments. Also, the sensitivity of one test strain decreased in more turbid juices. This difference was ascribed to the antioxidant power of the suspended solids, although data for the antioxidant power of the juices was not presented. *L. monocytogenes* irradiated *in vitro* was increasingly protected in solutions of increasing antioxidant power [43]. However, *S. enteritidis* suspended in commercial citrus juices of varying composition (orange vs. orange/tangerine, extra pulp vs. pulp free, regular vs. calcium enriched, etc.), and varying antioxidant power did not differ in D_{10} value following gamma irradiation [56]. The influence of natural and artificial antioxidants on radiation sensitivity is therefore an area to be addressed more fully.

11.11.2 SENSORY PROPERTIES OF IRRADIATED JUICES AND PULPS

A frequent added benefit of the application of ionizing radiation to fruits is the increase in juice yield during processing due to the softening of internal tissues. As in whole fruits, irradiation oxidizes a portion of juice ascorbic acid to the dehydro form; as in fruits, this conversion is expected to have little nutritional impact [134, 135]. Fetter et al. [143] irradiated a variety of commercially pasteurized juices to a maximum dose of 5 kGy and used taste panels to evaluate juice quality. The irradiated juices of orange, guava, tomato, red currant, black currant, apricot, peach, pear, and apple showed no reduction of flavor quality; the quality of similarly irradiated grape juice was reduced [143]. Chachin and Ogata [144] treated grape, apple, and orange juices with sterilizing (2 to 80 kGy) doses of gamma radiation. Loss of grape juice anthocyanin and orange juice beta-carotene was evident after 10 kGy, and was dose dependent up to 80 kGy. A dose of 5 kGy caused browning in apple juice after 5 kGy, and ascorbic acid was reduced in apple and orange juices. Addition of 0.01% propyl gallate reduced the negative effects of high-dose (10 kGy) irradiation on the quality of orange juice, but did not protect apple juice [144].

A later study of high-dose irradiation (10 kGy) of orange juice resulted in a similarly unacceptable degree of flavor degradation and browning; however, the addition of 0.1% sorbic acid before irradiation effectively eliminated loss of flavor and reduced browning [145]. Fan and Thayer [146] determined that irradiation of apple juice caused an initial reduction of the juice's brownness (A_{420}) and increased the antioxidant power of the juice. These effects were dose dependant up to 8.9 kGy. During subsequent storage of the juice at 5°C, the rate of browning of the irradiated juice was greater than that of nonirradiated juice, but after 16 d in storage, the irradiated juice was lighter in color. The impact of irradiation was influenced by processing temperature, but not by exclusion of oxygen from the juice or by the level of suspended matter in the juice [146]. Unpasteurized

Ionizing Radiation Processing of Fruits and Fruit Products

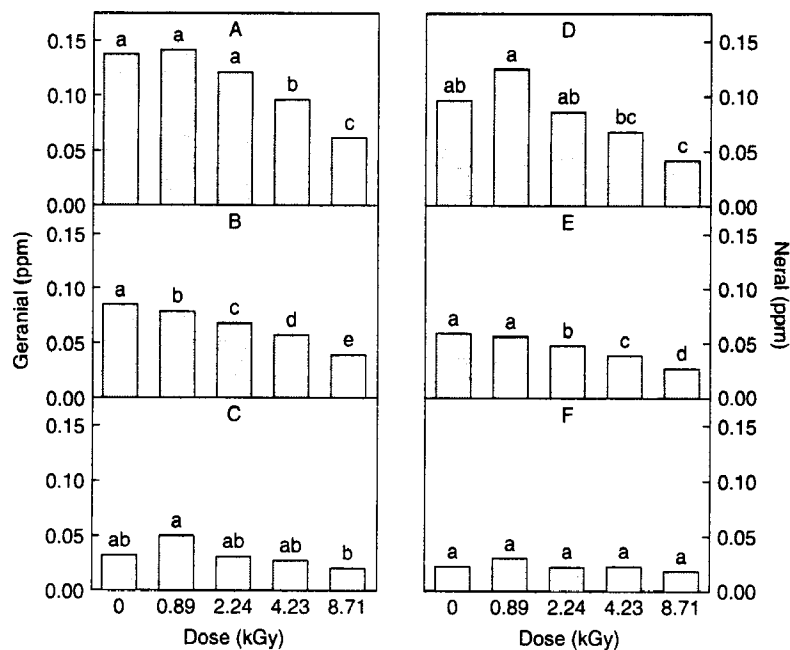


FIGURE 11.5 Concentration of geranial (A, B, and C) and neral (D, E, and F) in irradiated and nonirradiated orange juice. Juices were irradiated with 0.0, 0.89, 2.24, 4.23, or 8.71 kGy at 5°C, and stored at 7°C for up to 21 d. Volatile compounds were measured with gas chromatography after 1 d (A and D), 7 d (B and E), or 21 d (C and F). Bars with the same letter are not significantly different (LSD, $P < 0.05$). Comparisons were made within the same storage duration. (From Fan, X. and Gates, R.A., Degradation of monoterpenes in orange juice by gamma radiation, *J. Agric. Food Chem.*, 49(5), 2422–2426, 2001. With permission.)

apple cider irradiated to 3 kGy was identifiable, but not unacceptable, to untrained sensory panelists evaluating aroma; similar results were obtained with reconstituted and fresh orange juices (B.A. Niemira and X. Fan, unpublished data).

Niemira et al. [57] found no evident changes in the appearance or aroma of reconstituted orange juice irradiated to 2.5 kGy. Miller and McDonald [128] reported that the juice from irradiated grapefruit was acceptable after 0.3 kGy, but declined in quality at 0.6 kGy. Pickett et al. [142] reported increasing off-flavor in unpasteurized orange juice irradiated to 3 kGy, rendering the juice unpalatable. Spoto et al. [147] irradiated orange juice concentrate and determined the effect of storage time and temperature on juice quality and acceptability. In that study, the highest dose (up to 5 kGy) combined with 25°C storage resulted in loss of “orange” flavor and increase in “bitter,” “medicinal,” and “cooked” ratings by sensory panelists. Lower dose (2.5 kGy) and cooler storage (0 or 5°C) were proposed as an acceptable processing regimen. Fan and Gates [148] found that some aroma-related acyclic monoterpenes in orange juice were reduced immediately after irradiation; after refrigerated (7°C) storage for 21 d, there was no significant difference from the nonirradiated controls (Figure 11.5). A consumer test determined that irradiated apple cider was as acceptable or more acceptable than pasteurized apple cider [149].

The factors that may lead to undesirable flavors, aromas, or changes in appearance of irradiated juices are complex. While extensive studies into the effect of the variety and maturity of the source fruit on the radiation sensitivity of the resulting juice are lacking, it is known that juice composition is variety dependent [150]. In light of the evidence for varietal specificity in irradiated whole fruits and vegetables, the possibility of varietal specificity in juices with regard to negative irradiation effects cannot be discounted. As with whole fresh produce, temperature control during the irradiation process is important. The irradiation process can lead to generation of ozone in the headspace

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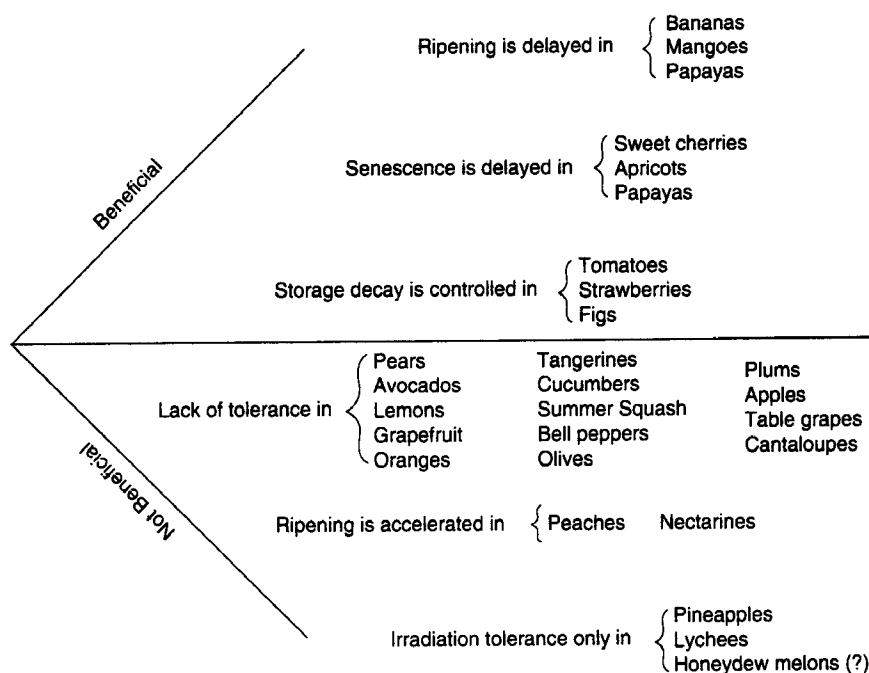


FIGURE 11.6 The response of 27 fruits to irradiation. (From Akamine, E.K. and Moy, J.H., Delay in postharvest ripening and senescence of fruits, in *Preservation of Food by Ionizing Radiation*, Vol. 3, Josephson, S.E. and Peterson, M.S. Eds., CRC Press, Boca Raton, FL, 1983, pp. 129–158. With permission.)

gas of sealed containers; consideration should be given to the possibility of secondary sensory effects from the ozone, rather than from the radiation directly. Another possible source of off-odors or flavors is the plastic used in the processing tests. A bag made of a plastic not suited for irradiation may cause unpalatable migration of radiolysis products. Another example is a fruit juice irradiated in a glass test tube with an unsuitable plastic cap or even a cap liner that is the source of a disagreeable odor or flavor. Any of these situations could lead to a mistaken conclusion regarding the suitability of irradiation for a particular product.

11.12 FRUITS CURRENTLY BEING IRRADIATED

A very large volume of information, occasionally contradictory, exists on specific applications of gamma irradiation to fruits [16, 17, 118, 151]. Figure 11.6 shows the response of a number of fruits to irradiation, as summarized by Akamine and Moy [151]. The results are often difficult to compare because of differences in conditions of treatment. A few representative examples are discussed below.

11.12.1 STRAWBERRY

Among fruits, strawberry is one of the most studied with respect to application of gamma irradiation. Market tests and commercial applications have been carried out in a number of countries. A dose of 2 kGy seems to be the optimal dose of irradiation in air for precooled, ripe strawberries. Success depends on cultivar: the firmer fruits of “Tioga” tolerated radiation better than the softer fruits of “Brighton” [152]. In 1992, commercial irradiation of strawberries was initiated in Florida. The fruits were irradiated at 0.3 to 1 kGy on pallets in air containing 10% carbon dioxide [153] at 50°F. The plastic wrapping was removed after 3 d. After 18 d of storage, the strawberries irradiated under

modified atmosphere were sweet, firm, slightly dark, free of mold, and still saleable, in contrast with control fruits held in air and fruits held under modified atmosphere for 3 d before unwrapping. According to several reports, marketing of the irradiated strawberries was quite successful [154].

11.12.2 MANGO

The D_{10} of 14 phytopathogenic fungi were evaluated in irradiated (0.5 to 1.5 kGy) mango pulp [58]. These ranged from 0.39 for *Aspergillus sydowii* and *Aspergillus ustus* to 1.11 for *Penicillium oxalicum*, confirming the relatively high radiation resistance of fungi. The D_{10} values were also determined in saline solution, and demonstrated some variance with the data obtained in mango pulp. The more complex medium caused a decrease in D_{10} for some fungi, e.g., *A. ustus* (55%), *Scopulariopsis brevicaulis* (55%), and *A. sydowii* (76%), and an increased D_{10} for others, e.g., *Penicillium brevicompactum* (147%), *Aspergillus flavus* (159%), and *P. oxicalum* (179%). The highest dose (1.5 kGy) reduced total aerobic bacteria count by ~1.8 logs, a difference that persisted throughout the study (50 d). Shelf life in irradiated (1 kGy) fruits was extended from 25 to 50 d in refrigerated storage. While the interiors of the fruit used in this study were sampled and found to be free of contamination, the variable effect of fruit pulp on the radiation resistance of resident microorganisms is clearly demonstrated. Pulp represents a combination of plant cell contents with extracellular fluid, found in the intercellular spaces; nevertheless, this observation should be considered in light of evidence of internalization of pathogenic bacteria into intercellular spaces [33].

Numerous reports indicate that mango preservation would greatly benefit from treatment with ionizing radiation. The summary by Akamine and Moy [151] shows the optimal dose to be 0.75 kGy for three-quarter ripe fruits at room temperature. Combination with mild heat treatment by hot water dip or vapor for 5 min at 50 to 55°C yields even better results [152]. A limiting factor is surface scalding [151]. The effects of irradiation depend on the degree of maturity. Scalding occurred at 0.25 kGy on the mature-green fruits, and tolerance increased with maturity. Refrigeration at 13°C reduced tolerance to scalding. The susceptibility to radiation injury varied greatly with the origin of the fruits, although, in general, mangoes are considered highly tolerant [15]. These various factors have to be balanced to give maximal shelf life based on delay of ripening and control of disease. A market test for irradiated mangoes in 1986 in Florida was highly successful.

Mangos irradiated to 1.5 kGy were as acceptable to a sensory panel as nonirradiated controls immediately after treatment based on odor, color, and overall appearance; irradiated fruit remained acceptable after 50 d of refrigerated storage, while nonirradiated controls became unacceptable after less than 30 d [58]. Mango is one of several fruits being researched as part of an IAEA cooperative research project on the application of irradiation to fruits, vegetables, and vegetable-derived products [35].

11.12.3 PAPAYA

Extensive research was carried out on gamma irradiation of papaya in Hawaii to overcome the strict quarantine regulations of the mainland against insects, particularly fruit flies. Papaya is considered relatively tolerant of radiation. An effective treatment is 0.75 kGy, in combination with a hot water dip at 48.9°C for 20 min [151, 152]. The combined treatment delayed ripening, as assessed visually and by measurement of firmness and decay [151]. Hot water dip alone accelerated ripening, while irradiation alone provided only slight control of decay. Little difference was reported between irradiated and fumigated papayas. Irradiated fruits were firmer and had a better flavor than ethylene dibromide-treated fruits. Hot water dip was preferable to vapor-heat treatment. Extension of shelf life was obtained even without refrigeration. The authors concluded in favor of the feasibility of gamma irradiation of papayas. Papayas are one of the fruits currently being irradiated commercially at an x-ray facility in Hilo, HI. These products receive a minimum of 0.25 kGy. Whereas fruit that is steam-treated must be picked green, the irradiated fruit is allowed to tree-ripen longer,

yielding a higher-quality fruit [88]. Irradiated papayas have quality and nutrition similar to heat-treated papayas [155].

11.13 COMBINATION TREATMENTS

As has been indicated earlier, achieving a 5-log reduction in bacterial population on fresh fruits and juices would require between 1.5 and 3.5 kGy [54–57]. The higher doses may be more than those many fruits would withstand from a quality standpoint. It is not surprising, then, that much interest has been expressed in combining lower doses of ionizing radiation with other processes to achieve a total reduction of 5 logs. In part, the goal of this research is to determine the extent to which two or more treatments applied together will deliver a greater kill than the sum of each individual treatments, i.e., synergistic effect vs. purely additive effect.

The actual physical and chemical mechanisms behind antimicrobial synergy are complex, and vary depending on the treatments being examined, but they generally follow the same underlying rationale. An appreciable percentage of the bacterial population that survives the first treatment is injured or weakened, rendering the surviving population more susceptible to the second treatment than a normal population; thus, the second treatment is more effective than the first. Depending on the treatments being applied, the sequence of treatments may be an important factor in obtaining synergistic kill vs. purely additive. In a study of irradiated *Y. enterocolitica*, Sommers and Bhaduri [48] showed that the plasmid-encoded virulence genes in the surviving population were damaged, rendering the surviving population less virulent. Damage to plasmid-encoded genes is therefore one possible mechanism by which irradiation may predispose a population to greater susceptibility to other antimicrobial treatments.

11.13.1 MILD HEAT

The combination of irradiation and mild heat (45 to 55°C) has been used successfully with papaya and mangoes for delayed ripening and control of anthracnose, even without refrigeration [151]. Heat could be supplied as a hot water dip or as water vapor. Storage under modified atmosphere brought additional benefits. Combination of a 12-min steam treatment and gamma irradiation (1 to 2 kGy) increased the shelf life of mango pulp to 270 d vs. 90 d for irradiated or nonsteamed, or 15 d for untreated mango pulp (nonsteamed and nonirradiated) [156]. The combination treatment had no undesirable effect on chemical or sensory properties of the mango pulp.

Fan et al. [157] determined that a combination of a warm water dip (47°C, 2 min) and low-dose irradiation (0.5 or 1 kGy) could effectively enhance the safety and quality of cut lettuce. Caution may be warranted, however, as the population dynamics of pathogens in storage, posttreatment, are not fully understood. Lettuce was treated with a combination of warm (47°C) water and chlorine (100 µg/ml), and inoculated either before or after these treatments with *L. monocytogenes* and *E. coli* O157:H7. After an initial reduction, these pathogens rebounded and grew extensively on lettuce stored at abusive temperatures (10°C). Growth was not observed in lettuce treated with cold (1°C) water plus chlorine [158]. Though this study did not include irradiation as a treatment, it illustrates the importance of proper controls over every processing step, particularly when the effect of interacting treatments is not fully understood.

11.13.2 MODIFIED ATMOSPHERE PACKAGING (MAP)

Low-dose irradiation combined with modified atmosphere is increasingly considered for control of microorganisms and delayed ripening. As stated above, reduced oxygen content in the atmosphere limits the efficiency of irradiation because less activated oxygen is available. An interesting alternative is irradiation in air enriched with carbon dioxide. Carbon dioxide is fungistatic at levels above 12%, a concentration beyond the tolerance of most plant tissues (strawberries are an excep-

tion). Couture and Willemot [159] showed the synergistic action of the combined stresses of gamma radiation and high carbon dioxide for control of mold development on strawberries. Irradiation at the low dose of 0.3 kGy, combined with storage in air containing 10% carbon dioxide, delayed ripening, as assessed by anthocyanin content and mold development. The method has since been adapted commercially in Florida [153].

MAP of fruits involves equilibrating the respiratory carbon dioxide production and oxygen depletion by the plant material with the film permeability in order to maintain an adequate composition of the atmosphere (oxygen and carbon dioxide concentrations) within the limits of tolerance of the fruits inside the package. With irradiated fruits, the temporary increase in the rate of respiration due to the initial wound response has to be taken into account. The technology can be applied in several ways, e.g., irradiation of a wrapped pallet, of small individual containers, or of small perforated containers under a common wrapping on a pallet. Accumulation of carbon dioxide and depletion of oxygen may lead to fermentation and development of off-odors and off-flavors. Perforated, microporous, or semipermeable films may resolve this problem. Preirradiation flushing of the package with a low-oxygen gas mixture would reduce the undesirable effects of irradiation on the produce, e.g., loss of vitamin C, and on the packaging material, e.g., development of undesirable flavor and odor; however, this reduces the efficacy of the process. Sealed packaging acts as a water barrier and reduces weight loss. The integrity of the seal should resist the irradiation treatment in order to maintain the internal modified atmosphere and to prevent microbial recontamination. A study of lettuce and cabbage packed in a high-nitrogen atmosphere showed an extension of shelf life but no inhibitory effect on resident microflora, prompting the authors to caution against the potential for growth of pathogens on vegetables so packaged [160]. Irradiation of this type of packaged product would seem to be a valid means of addressing this concern.

Combination of irradiation with MAP has shown success with irradiated packaged meats [59], and is an area of active research on fruits and vegetables [35, X. Fan, personal communication]. Couture and Willemot [159] and Brecht et al. [161] showed beneficial effects of low-dose irradiation and storage under modified atmosphere for strawberries. Brecht et al. [161] obtained reduced decay incidence of *Rhizopus stolonifer* and *Botrytis cinerea* on strawberries packaged under 7% oxygen and 20% carbon dioxide and irradiated at 1 kGy. Grandison [99] reported a reduced rate of respiration and a delay of up to 3 d in ripening for bananas exposed to 0.1 to 0.3 kGy in an electron accelerator. Higher doses caused browning of the skin, splitting of the fruits, and loss of peel texture and vitamin C. Combination of irradiation with MAP under a 25- μ m low density polyethylene (LDPE) film extended the shelf life, although not synergistically. MAP did not alleviate the disorders caused by electron beam irradiation to avocados. After irradiation under MAP, total aerobic counts were reduced by ~3 logs on iceberg lettuce [137] and by ~1.5 logs on romaine lettuce [96] by doses of 0.19 kGy and 0.35 kGy, respectively. Rate of spoilage and shelf life was reported to be unchanged for iceberg lettuce, but the shelf life of romaine lettuce was extended from 14 to 18 d to the length of the study, 22 d. MAP reduced total aerobic plate counts by ~2 logs on shredded carrots irradiated to 0.45 kGy, a difference in microbial population that persisted throughout a 9-d storage evaluation [162]. Many minimally processed fruits and vegetables, such as prepared salad mixes, incorporate MAP as a means to control spoilage and extend shelf life. The precise gas permeabilities of the plastics used in the packages are of critical importance for these products. Especially with regard to fruits and vegetables, which respire during storage, studies of MAP plus irradiation must take into account the effects of irradiation on the material used to hold the product.

11.14 IRRADIATION OF PACKAGING MATERIAL

Like other foods treated by irradiation, fruits are prepackaged to facilitate handling and to prevent recontamination. Irradiation of fruits involves constraints on the types of packaging materials

suitable for this technology. The combination of irradiation and MAP brings additional constraints on the suitability of the packaging material. Irradiation may affect the characteristics of materials and packaging performance. It is therefore important to know these effects as they will influence the choice of a packaging compatible with the requirements of the agri-food industry. The functional properties of the material should be preserved. The toxicological and sensory aspects should also be addressed. Although the effects of irradiation are generally minimal, i.e., below 10 kGy, volatiles may be generated during the treatment. They may be transferred to the food and may produce unpleasant flavors. Migration of other compounds through contact while packaging food must be monitored to insure that the food is chemically harmless.

Selection criteria for packaging materials to be used with irradiation include sealability, resistance to cracking and delamination, protection against recontamination, barrier to oxygen and humidity, and chemical inertness. Traditional packaging materials such as metal, glass, and paper are relatively resistant to ionizing radiation. Metal and glass are essentially impervious to the effects of irradiation, although with high doses, glass can become somewhat browned. Paper and paper products such as pressboard, cardboard, etc., can lose mechanical strength due to the breakdown in cellulose at the microscopic level. However, the low water content of these products greatly reduces the sensitivity of the cellulose in paper vs. that in fruit or vegetables. Paper and cardboard are used mainly for support packaging.

Plastics are the type of packaging that most frequently comes into contact with the food treated with ionizing radiation. Plastics have historically been basic resin polymers to which additives (e.g., stabilizers, antistatic and antislip agents, and plasticizers) have been added. Radiation acts on both the polymers and additives. Increasingly, the plastic films used for packaging are laminates, with two or more layers of dissimilar plastics combined to provide the desired mixture of strength, durability, gas permeability, ability to take inks, etc. Determining the effects on the packaging material is thus made more difficult as each plastic component of the laminate has a unique response to irradiation, and this response may or may not be the same for the plastic as part of a laminate as it would be for the plastic standing alone. Also, some specialty laminates can incorporate a metal foil layer, adding another layer of complexity.

A relatively recent development in packaging technology has been the incorporation of preservatives and antimicrobial compounds within the plastic, designed to suppress the growth of spoilage organisms or pathogens [163]. These are of increasing interest to food processors and food safety professionals. For use in food products to be sold in the U.S., these compounds must be cleared by the U.S. FDA.

11.14.1 TYPES OF PLASTICS USED IN FRUIT PACKAGING

Fresh fruits are usually packaged in plastics of high or intermediate permeability (Table 11.2). The most commonly used materials are LDPE, polypropylene (PP), polyvinyl chloride (PVC), and ethylene vinylacetate (EVA). Seasonal trade requires punnets made of woven chip or thermoformed polystyrene (PS) with LDPE or PVC cling wrap. Small polyethylene terephthalate PET trays are also becoming popular. Juice-absorbing pads can be used to soak residual liquid from the fruit or prepackaging rinses. Properties of PET are well preserved during irradiation [164]. In the U.S., PS films have been approved for irradiation; however, the PS foam trays have to be PE-coated, which increases packaging cost. Single-layer plastic films commonly used for fruit packaging, including LDPE, PP, PVC, polyvinylidene chloride (PVDC), and PET, released odors after irradiation at the low doses used for fruits [165]. At high doses, cellulosic materials, such as paper and board, lose mechanical strength. At the doses typically used with fruits, this effect is minimal and may be offset by using PE or foils as a laminate for support [166]. Adhesives used in absorbing pads are not permitted for irradiation in the U.S. and Canada. Pads with a mechanical link between paper and polyethylene (PE) layers would be a solution. PP and PVC are not recommended for use with irradiated fruits for the reasons discussed earlier. Several suitable materials have not yet received

TABLE 11.2
Suitability of Plastic Materials for Fruit Irradiation

Polymer	Use	Suitability
Polyethylene (PE)	Pallet shrink packaging, bags, lids coatings, wrap films	Good
Polypropylene (PP)	Bags	Mediocre
Ethylene vinyl alcohol (EVA)	Coatings, MAP, frozen fruits, sealing	Good
Polyvinyl chloride (PVC)	MAP, overwrapping	Poor
Polyethylene tetraptalate (PET)	Bags	Good
Polystyrene (PS)	Trays, films	Excellent
Polyvinylidene chloride (PVDC)	Barrier layer	Good
Nylon-8	Bags	Excellent
Nylon-11	Bags	Good
Cellulose	Bags, overwrapping	Mediocre

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tials.

Source: Willemot, C.M., Maronette and Deschenes, L., Ionizing radiation for preservation of fruits. in *Processing Fruits: Science and Technology*. Vol 1: *Biology, Principles and Applications*. Somogyi, L.P. and Ramaswamy, H.P., Eds., Technomic Publishing Lancaster, PA, 1996, pp. 221–260, chap. 9.

clearance for irradiation. Because of the complexity of the problem, the applications have to be evaluated individually.

Dried fruits irradiated for insect disinfestation are stored for long periods. The durability of the irradiated material has to be tested. Barrier films used with dried fruits are typically multilayers of polyolefins, EVA, PVDC, or nylon. Oxygen barrier films reduce radiation-induced vitamin C loss [166]; however, films laminated with adhesives have not yet been cleared for irradiation.

At the present time, no packaging material can be considered suitable for irradiation of fruits unless appropriate migration tests have been performed. Even with approved plastic films, off-odors may be generated; however, as off-odors are typically the result of radiation-induced rancidity of fats, this risk is low in most fruits because of the low fat content (2). The main parameters for the choice of a packaging material before fruit irradiation are the FDA list and the specific requirements of the fruits.

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11.14.2 EFFECTS OF IRRADIATION ON PACKAGING PLASTICS

The effects of the ionizing radiation on polymers are mainly cross-linking and cleavage. The two effects can occur simultaneously, and the relative importance of each depends on the material and on the conditions under which the treatment is carried out [167]. Cross-linkage, the formation of bonds between adjacent chains, generates a very resistant three-dimensional network. The extent to which cross-linkage occurs between the layers of plastic laminates will depend on the plastics involved. Cleavage refers to reduction in chain length and, thus, to a loss of mechanical strength and an increase in porosity of the material. In general, the low doses applied to fruits cause only insignificant modifications of the physical properties of plastics; however, some materials are more susceptible to ionizing radiation and are not recommended for irradiation of prepackaged food.

Mass transfer from packaging material to food after irradiation may involve migration, absorption, and permeation. Research has demonstrated that these three phenomena can be affected by irradiation.

Migration refers to the transfer of low molecular weight molecules to the food and is of importance for toxicology and organoleptic quality. The plastics used for fruit packaging are mostly very high molecular weight thermoplastics derived from ethylene. They are chemically inert and have a low solubility in water and oil; therefore, migration concerns mainly low molecular weight additives and their products generated by irradiation. Migration rates have been shown to be affected

by radiation [164]. The effect is more important with gamma rays than with accelerated electrons. Irradiation of plasticized PVC film causes migration of HCl and tainting of food by other low-molecular-weight molecules [166]. Kilcast also showed migration from irradiated PET and high-impact polystyrene (HPS) trays. He suggested the use of expanded PS. No taint was observed from PE-based films, while PP is known to be very susceptible to irradiation [168]. Bourges et al. [168] showed that oxidation products of antioxidant additives produced at doses below 10 kGy could migrate into food-simulating solvents. A study (gamma and e-beam) of two semirigid copolyesters showed that 5 kGy had no effect, and that 25 or 50 kGy had no significant effect on migration of nonvolatiles into food-simulating solvents [169]. A later study (gamma) of an amorphous nylon polymer using the same doses (5, 25, and 50 kGy) showed the generation of volatiles but not of nonvolatile compounds [170]. No new compounds were detected in that study.

Comparatively little is known at the present time about toxicity of degradation products of many other additives. The extent to which the concentration, efficacy, or migration of these antimicrobial compounds (or any of their radiolytic products) are affected by irradiation is essentially unexplored [163].

Permeation, the transfer of molecules through the packaging material, is only slightly affected by irradiation below 10 kGy [165]. This aspect should, however, be considered in the context of MAP [171].

Absorption of volatiles and other constituents of food by packaging material can be important. Scalping of terpenes that contribute to the aroma of citrus fruits by packaging material [172] is a serious problem for the fruit juice industry. Absorption has been only slightly studied in the context of food irradiation.

11.14.3 REGULATION OF PACKAGING MATERIALS FOR USE WITH IRRADIATED PRODUCTS

The *Codex General Standard for Irradiated Foods* [173] established worldwide standards for packaging of irradiated food. These guidelines are very general and offer little precision with respect to the types of materials and additives suitable for prepackaging of food to be irradiated. The U.S. is the only country with a list of packaging materials approved for irradiation (Table 11.3). Some of these plastics may be amended with various adjuvants, including preservatives, etc. (Table 11.4). The FDA list draws on data from research carried out by the U.S. Army on irradiation-sterilized food packs, as well as data from more recent research. The regulation covers only plain films or laminates, presently used in food packaging. Juice-absorbing pads in film-covered trays often contain adhesives between plastic and paperboard layers. These pads and trays such as the popular uncoated polystyrene foam trays are not approved for irradiation, although polystyrene films may be irradiated.

In Canada, no such list of packaging materials approved for irradiation has been established. The standards are intended to serve only as guidelines. Whatever may be the material or the process, it must not constitute a health hazard. Until August 1988, there were no specific approvals for packaging materials. Since then, permission has been granted for Cryovac films and for bags used in the shrink- and vacuum-packaging of refrigerated products [174]. In Canada and the U.S., a clearance petition should be presented to the authorities for plastic materials that are not mentioned in the FDA list and that come in contact with the food during irradiation. Several materials that are good candidates for packaging of irradiated fruits cannot currently be used. These materials await a concerted effort of industry groups to petition the appropriate governmental regulatory bodies for approval.

11.15 COST-BENEFIT OF FRUIT IRRADIATION

The beneficial effects of irradiation for fruit disinfestation, reduction of spoilage, and extension of shelf life offer potential for significant cost savings to the food industry. One of the major concerns

TABLE 11.3

U.S. Code of Federal Regulations 21CFR179.45: Packaging Materials Approved for Irradiated Foods

Material	Maximum Dose
Nitrocellulose-coated or vinylidene chloride copolymer-coated cellophane	10 kGy
Glassine paper	10 kGy
Wax-coated paperboard	10 kGy
Films of polyolefin or polyethylene terephthalate. These may contain:	10 kGy
1. Sodium citrate, sodium lauryl sulfate, polyvinyl chloride ^a	
2. Coatings comprising a vinylidene chloride copolymer containing a minimum of 85% vinylidene chloride with one or more of the following comonomers: Acrylic acid, acrylonitrile, itaconic acid, methyl acrylate, and methyl methacrylate	
Kraft paper (only as a container for flour)	0.5 kGy
Polystyrene film	10 kGy
Rubber hydrochloride film	10 kGy
Vinylidene chloride–vinyl chloride copolymer film	10 kGy
Nylon 11	10 kGy
Ethylene–vinyl acetate copolymers	30 kGy
Vegetable parchments	60 kGy
Polyethylene film ^a	60 kGy
Polyethylene terephthalate film ^a	60 kGy
Nylon 6 films ^a	60 kGy
Vinyl chloride–vinyl acetate copolymer film ^a	60 kGy
Acrylonitrile copolymers ^a	60 kGy

^a This material may be amended with additional materials, listed in Table 11.3.

AU: Please verify the table number mentioned.

TABLE 11.4

U.S. Code of Federal Regulations 21CFR179.45: Adjuvants and Amendments Approved for Incorporation into Certain Packaging Materials Approved for Irradiated Foods

Adjuvant/Amendment	Limit (By Weight of Polymer)
Amides of erucic, linoleic, oleic, palmitic, and stearic acid	1%
BHA (butylated hydroxyanisole)	1%
BHT (butylated hydroxytoluene)	1%
Calcium and sodium propionates	1%
Petroleum wax	1%
Mineral oil	1%
Stearates of aluminum, calcium, magnesium, potassium, and sodium	1%
Triethylene glycol	1%
Polypropylene, noncrystalline	2%

of any potential user is whether the process is economically viable. Although irradiation plant capacity and processor commitment to the technology are growing at perhaps the fastest rate since irradiation was first developed over 40 years ago, the economic feasibility of irradiation has to be examined for each individual application. Also, the cost of this innovative technology has to be compared with other existing processing technologies [175].

The cost of gamma irradiation is similar to irradiation using an electron accelerator. The latter source is not likely to be used with fruits because of the low penetration capacity of the electron beam; however, electron accelerator-generated x rays, with significant improvement in the x-ray yield, may become an interesting alternative for bulk irradiation of fruits.

Irradiators are costly to install, and a substantial capital investment is needed [131]. The main factors contributing to the initial cost of any irradiation facility are the source (radionuclides or electron accelerator equipment), the shielding, and the product conveyer system. This important capital investment has delayed the development of food irradiation. Lack of food irradiation capacity means that irradiated food cannot be in the marketplace, and it has no opportunity to gain market share. Without market share, there is insufficient economic incentive for the capital outlay of a new irradiation plant.

The main operating costs of a gamma irradiator include depreciation and replacement of the source, while maintenance of the electronics and the electricity consumption are more significant factors for e-beam and x-ray irradiators. Urbain [13] concluded that the operating costs for food irradiation were in the range of commercial feasibility. The cost of irradiating foods was estimated between U.S. \$0.02 and \$0.04 per kg [176]. Giddings [177] estimated the processing cost at \$0.05 for a throughput of 10 million kg a year. A study by Fundacion Chili in 1985 evaluated the cost of disinfesting fruits by gamma irradiation at \$0.025 per kilogram for \$1 million capital investment in a batch-type cobalt-60 facility [178].

Cost estimates are dependent on the throughput capacity of the irradiation plant, the dose delivered, the distance the produce must travel to and from the irradiator, the distance between the plant and the markets being served, etc. Ancillary market benefits of irradiation (reduction of storage losses, premium prices commanded by "specialty" markets, etc.), may be offset by ancillary market drawbacks (necessity for increased public education or outreach spending, potential for increased regulatory oversight, etc.). These complex factors make an accurate cost-benefit analysis of food irradiation difficult. There are few examples of large-scale commercialization from which to gain insight, but the recent increase in irradiation of ground meats and Hawaiian fruits is expected to allow for real-world data to be applied to these calculations.

Kunstadt and Steeves [179] discussed in detail the economics of fruit disinfestation by irradiation and the effects of various parameters on the unit-processing cost. They presented a detailed analysis of the costs incurred for different types of cobalt-60 irradiators; this analysis remains instructive as x-ray treatment of fruits becomes more prominent in the marketplace. The influence of dose, packing density, and volume of operation was calculated. As expected, unit-processing costs decrease rapidly with increased throughput. Therefore, an economically successful irradiator has to operate at a level exceeding the minimum economic volume of production. It has to be designed for a realistic throughput. The unit-processing cost increases linearly with increasing dose of irradiation; it also depends on processing time. The linearly increasing cost with decreasing packing density is related to the efficiency of utilization of the source. Similar trends were observed with different types of irradiators. According to these calculations, the unit-processing cost of an irradiator designed for throughput of 60 million kg/year at a density of 0.4 g/cm³ and a dose of 0.15 kGy would vary by less than \$0.01 for a throughput of 250,000 t/year to \$0.107 per kg for 10,000 t.

11.16 THE FUTURE OF IRRADIATION PROCESSING OF FRUITS

Treatment of fruits by irradiation is increasingly being recognized as an effective method for reducing postharvest losses, ensuring hygienic quality of produce and facilitating international trade of specific commodities of tropical origin; however, worldwide, more than 80% of food irradiation is applied for the disinfection of spices, herbs, and dehydrated seasonings. The industry has shown

widespread interest in this technology, has provided quality control through the necessary permissions that were granted, the quality of the treated produce was high enough to offset the price increase, the cost per unit was acceptable to the consumer, and no mandatory negative labeling would discourage purchasing.

Loaharanu [154] reported on several successful market trials, mainly for fruits, in the 1980s. In 1986, 2 t of mangos were irradiated at 1 kGy in Puerto Rico and flown to Miami. In 1987, Hawaiian papayas were shipped to Los Angeles and irradiated at a low dose. In both trials, the fruits were well received by consumers. In Lyon, France, in 1988, strawberries irradiated at 2 kGy sold at a slightly higher price than nonirradiated fruits. The consumers preferred the irradiated strawberries for their better quality. Since the opening of the \$6.8 million Vindicator irradiator at Tampa, FL, in January 1992, various fruits have been irradiated and sold successfully in Florida and Chicago. The Hawaiian fruits now being treated with x-rays are reportedly selling well in their target markets. These fruits include papaya, avocado, rambutan, lychee, and star fruit. The increasing capacity of food irradiation facilities in the mainland U.S. suggests that as the processors seek to maintain their plants at or near full capacity, the range of products being treated may expand from ground and frozen meats to include more fruits and vegetables. Ongoing research and progress concerning the technical aspects of irradiation of fruits should further develop the use of this technology. Choice of cultivars, harvesting conditions, degree of maturity, combination treatments, suitable packaging, and minimum doses required for the desired benefits are among the parameters that will lead toward better quality and longer shelf life of irradiated fruits.

The phasing out of several fumigants used for disinfestation is likely to encourage the use of irradiation. Following the U.S. ban on ethylene dibromide, many producers turned to hot water dips to disinfest mangoes and papayas; however, this treatment may cause physiological disorders to the fruits, particularly discoloration and internal browning. Irradiation is an alternative process of considerable interest because it controls microbial development and sterilizes or kills insect pests without affecting fruit "freshness." It may become a major technique for meeting the quarantine requirements of several countries [180], including the U.S., with respect to disinfestation of fruits against insects such as the Mediterranean fruit fly. Developing countries will particularly benefit from the technology. Their often warm and humid climates accelerate ripening and decay of produce, often causing extensive losses. Most produce of tropical origin, e.g., mango and papaya, are chill sensitive. Irradiation is an alternative to low-temperature storage for preservation of some of these products. Developing countries are at great distances from their main industrialized markets, and irradiation would help maintain quality and safety of produce during transportation. The increase in consumption of fruits of tropical origin from developing countries, combined with the demand for safe, nutritious, and convenient food in industrialized countries will likely contribute to the expansion of the use of this technology. This can only serve to increase the trade in tropical fruits, thus benefiting the exporting and importing nations.

Food-borne illness resulting from contaminated produce and juices has encouraged research into the use of irradiation of fruits for the purpose of improving the safety of these products. Sanitization, a goal that has a resonance with consumers, has joined the traditional goals of disinfestation, delay of ripening, and shelf-life extension as stated aims of produce irradiation. Ultimately, the consumer will benefit from the presence in the marketplace of a greater variety of fruits, including tropical fruits, year-round, that are fresher, longer-lived, healthier, and safer than before.

ACKNOWLEDGMENTS

The authors would like to acknowledge the thoughtful reviews of J.L. Alford and J.S. Novak, and express grateful appreciation to K. Lonczynski for technical assistance in the preparation of this manuscript.

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Ionizing Radiation Processing of Fruits and Fruit Products

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